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CLIMATIC IMPACT ASSESSMENT PROGRAM

HOP ON COMPUTATIONAL MODELING OF THE ATMOSPHERE

ASILOMAR, PACIFIC GROVE, CALIFORNIA

11-14 April 1972

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Robert L. Underwood, Editor

July 1972

Final Proceedings

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16. Abstract

On 11-14 April 1972, the Department of Transportation sponsored a workshop on computational modeling of the atmosphere pertinent to assessment of possible climatic changes that might result from the projected 1985-1990 world high-altitude aircraft fleet. These proceedings record the deliberations of the two separate workshop panels: the fluid dynamics modeling panel and the chemistry panel.

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I. INTRODUCTION

The Department of Transportation (DOT) was created by the Congress for the purpose of addressing the nation's transportation as a single system, for thinking through how that system can grow to a better balance to serve all the people, while providing comprehensive protection to the environment in which it serves; and for doing those things the Federal Government ought to do to help achieve those goals.

Transportation R&D includes not only developing the technology of new systems but the careful, highly technical assessment of the effects that technology may have, with provision for fully controlling those effects—noise, emissions, visual pollution, violation of our precious wilderness, and all others.

The Department of Transportation takes very seriously the responsibility for assessing preveniently the effects that proposed transportation systems may have—for preventing ill effects when this can be done, and for preventing deployment of the systems themselves whenever it cannot.

A salient case in point is the possible use of supersonic transports as one component of our transportation system. In many ways the potential adverse effects of the SST are similar to those of other transportation systems.

But in one respect the SST just might present a unique problem. It has been suggested that the high-volume operation of a large SST fleet might produce a sensible change in the climate.[1/]

The United States has been a source of extended discussion regarding potential atmospheric effects from large-scale operation of SST's in the stratosphere. U.S. Congressional hearings, scientific panels of the U.S. Commerce and Transportation Departments; non-governmental scientific panels such as the Massachusetts Institute of Technology (MIT)-sponsored 1970 Summer Study of Critical Environmental Problems (SCEP Study), and individual scientists have reviewed facts, theoretical possibilities, opinions and speculations regarding the potential effects of engine emissions—from jet aircraft operating in the stratosphere—upon climate and weather, upon the temperature at the earth's surface, and upon man himself. It is widely regarded that an uneasiness over the lack of firm scientific answers to environmental uncertainties and speculations played a major role in the decision of the U.S. Congress to discontinue further development of the U.S. SST.

It is generally recognized that the effects of a few aircraft operating in the stratosphere are not significant. The scientific issues center on the large amounts of fuel which will be burned in the stratosphere when several hundred supersonic aircraft are in regular commercial service, thereby injecting into the stratosphere, over the course of a year, amounts of exhaust emissions which may be comparable to naturally injected amounts of the same constituents. While daily rates of injection of exhaust emissions may be relatively small, the high thermal stability of the stratosphere reduces the rate of vertical mixing and is generally expected to result in the slow accumulation of exhaust products and in characteristic removal times on the order of a year or so. The thermal stability is due to the radiative properties and photochemical reactions of the trace gases ozone, water vapor, and carbon dioxide.

[1/] These introductory paragraphs are excerpted from an address by the Honorable Robert H. Cannon, Jr., Assistant Secretary of Transportation, to the American Meteorological Society, San Francisco, 12 January 1971. The complete text is Appendix A to this document.

Although scientists in many countries have been studying the composition and meteorology of the stratosphere for a number of years and have greatly increased the depth of knowledge of the stratosphere's role in atmospheric processes, the recent discussion of the effects of SST engine emissions has delineated specific areas where uncertainties in present knowledge need resolution. For example, the natural concentrations, variability in time and space, sources, sinks, and removal rates for a number of trace gases, such as NO, H_2O, CO, and others, should be investigated in sufficient depth to permit comparison with injection rates from high-altitude aircraft.

The amounts of carbon dioxide and water vapor produced by jet aircraft engines can be estimated with sufficient accuracy for study of their effects from knowledge of the fuel flow rates under cruise conditions. Details of the engine design and operating conditions may affect strongly the amounts of such engine emissions as hydrocarbons (H_xC_y), nitric oxide (NO), carbon monoxide (CO), and the size distribution and composition of particulates. These engine emissions have been measured for many engines under sea level static test conditions, but rarely under actual or simulated cruise conditions.

During the debate on the U.S. SST, the U.S. Government took the position, expressed by Secretary of Transportation John A. Volpe, that environmental effects of the aircraft would have to be carefully assessed. In 1970, the Department of Transportation proposed to carry out the necessary research program. The research program, called the Climatic Impact Assessment Program (CIAP), was initiated by the Department of Transportation in FY 1971 and given substantial new impetus in FY 1972.

The Climatic Impact Assessment Program (CIAP) is to assess by 1974 the environmental and meteorological effects of the projected 1985-1990 world high-altitude aircraft fleet, including subsonic and supersonic vehicles. It is designed to provide a necessary part of the base of knowledge required to support Federal policy decisions concerning the operation of supersonic aircraft over territory under the jurisdiction of the United States. It addresses

the complex interactions between the engine emissions exhausted into the upper atmosphere, the natural composition of the stratosphere, and the dynamic processes of the atmosphere. It examines the effects of projected changes in the upper atmosphere upon the climate close to the earth's surface.

Conduct of the CIAP is the responsibility of Dr. Robert H. Cannon, Jr., Assistant Secretary of Transportation for Systems Development and Technology. Dr. Alan J. Grobecker, Manager of the CIAP, reports directly to Assistant Secretary Cannon. Figures 1.1 and 1.2 show the organization of the Department of Transportation and the CIAP management structure.

To meet the objectives of the Climatic Impact Assessment Program by the required data of 1974, the program is proceeding in the six steps listed below:

1. Determine nature of the stratosphere—constituents, properties, motions
2. Determine nature of the propulsion effluent—engine emissions testing, projections of routes and frequency
3. Estimate perturbed stratosphere—changes in height distribution of constituent densities, changes in energy transport
4. Estimate perturbed troposphere—changes in UV flux at the earth's surface, changes in heating, temperature, winds, precipitation
5. Determine physiological/biological/botanical impacts of man, animals, plants
6. Assess economic impacts.

Computational modeling is essential to the accomplishment of the first four steps in the program.

DEPARTMENT OF TRANSPORTATION

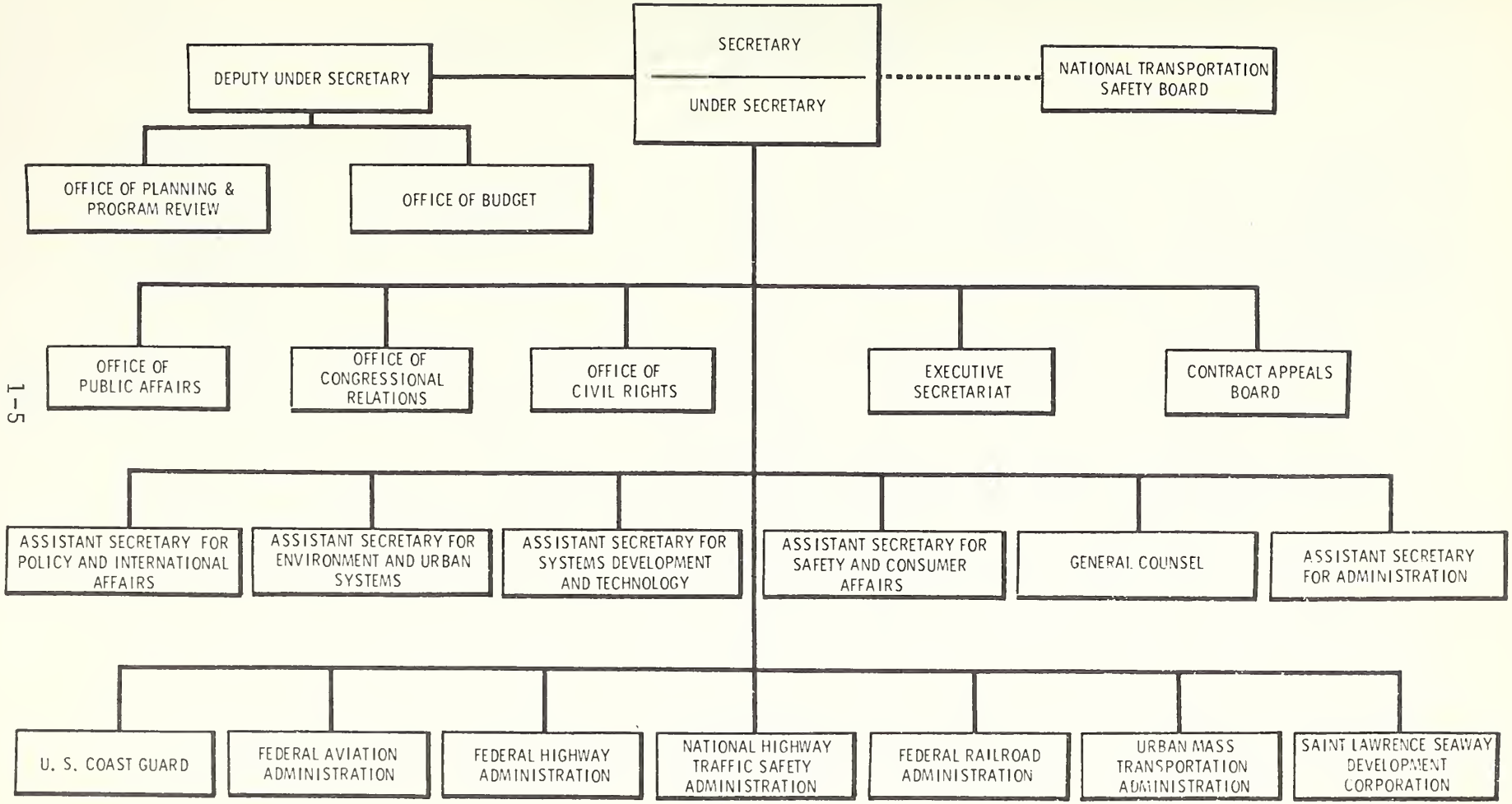

FIGURE 1.1. ORGANIZATION OF THE DEPARTMENT OF TRANSPORTATION

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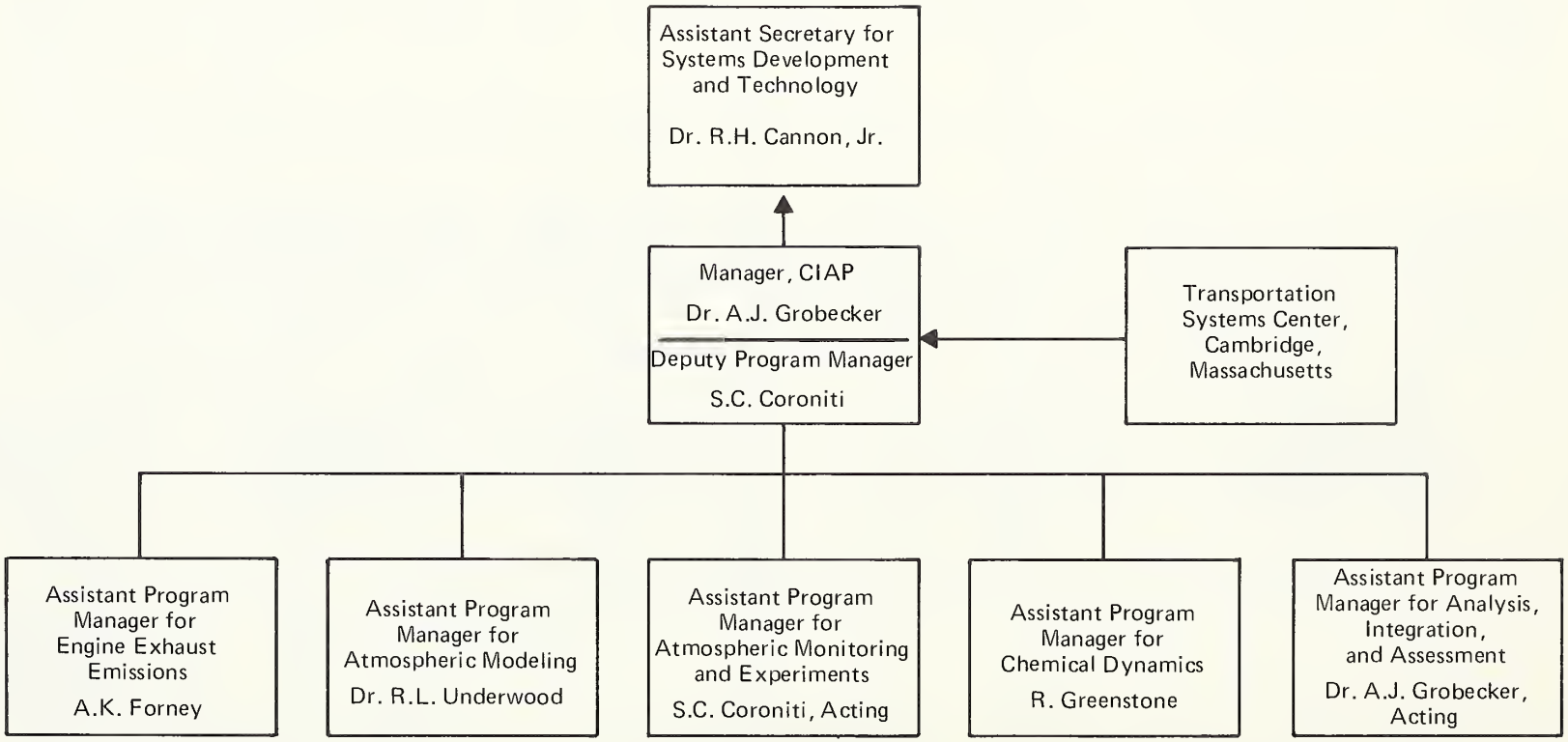

FIGURE 1.2. THE CIAP MANAGEMENT STRUCTURE

The steps in the program and the way the various steps relate to each other are described in Appendix B. A set of important technological questions to be addressed by the CIAP program are also discussed in Appendix B, and among these are three dealing with simulation and computer modeling:

- "What considerations must be made in the simulation of the stratosphere by computer?"
- "What are the expected limitations of the stratospheric simulation to be done in the next two years?"
- "What abridgements of a complete model of the stratosphere/troposphere/ocean, such as that planned for future development by NOAA's Geophysical Fluid Dynamics Laboratory, may seem appropriate to derive answers desired by CIAP?"

To address these questions, a Computational Modeling Workshop was sponsored by the U.S. Department of Transportation and held at Asilomar, Pacific Grove, California, April 11-14, 1972. The workshop was organized by Dr. R. L. Underwood of the DOT and Professor S. V. Venkateswaran of UCLA.

The workshop was divided into two separate panels, a Chemical Modeling Panel chaired by Dr. Halstead Harrison of the University of Washington and a Fluid Dynamics Modeling Panel chaired by Professor Yale Mintz of UCLA. Appendix C is a complete list of participants and their affiliations. As a prime workshop objective, each panel was asked to review ongoing and planned activities of its members pertinent to the CIAP objectives.

A second objective of the modeling workshop was to encourage interaction between the chemical and fluid-dynamic modelers. Traditionally, the chemical modelers and the fluid-dynamic modelers have worked independently and have approached the problem of modeling the atmosphere from different points of view. The chemical modelers, for instance, have concentrated their

efforts on the mesosphere and thermosphere while the fluid-dynamic modelers have been more concerned with the troposphere. Coordinated contributions from both scientific disciplines, however, are required to accurately model the stratosphere.

A third objective of the modeling workshop was to determine which, if any, of the current models of the atmosphere afford the most promising approach and are most likely to contribute to the CIAP program within the 1974 time constraint. If none of the current models were deemed appropriate to the CIAP task, it was hoped that new models directed specifically at the CIAP problem could be defined for development.

The following sections of this report present (a) the fluid dynamics panel report, which consists of a review of current modeling programs; (b) the chemical modeling panel report, which consists primarily of an inventory of possible important chemical reactions and an examination of the possible chemical influences and effects; and (c) a concluding section in which the achievements of the Asilomar workshop are summarized and evaluated.

II. REPORT OF THE FLUID DYNAMICS PANEL

Y. Mintz, Chairman

OVERVIEW AND SUMMARY

The Fluid Dynamics Panel reviewed in detail ongoing and planned activities of its members pertinent to the CIAP objectives. To a limited extent, note was taken of duplication in these activities and of gaps that would have to be filled if the CIAP objectives are to be met. Three distinct classes of models were considered:

- Models for the chemically reacting wake of high-flying aircraft which predict changes occurring in engine exhaust products in the flow field behind the aircraft to distances at which the influence of stratospheric dynamics becomes significant or dominant in comparison with aerodynamically induced flow perturbations
- Transport and dispersion models which describe how the exhaust emissions products are removed by the natural winds of the stratosphere from the region of the aircraft flight track and enter the global circulation

- Global-scale circulation models which are needed to relate any changes in the meteorology or physics of the stratosphere to atmospheric characteristics at or near the earth's surface.

The fundamentals of turbulence for application to both fluid flow and chemical kinetics were addressed. Dr. Hilst pointed out the effect of turbulence on chemical reactions with the conclusion that in the stratosphere turbulence will frequently affect the magnitude of effective reaction rates. In addition, Dr. Hilst indicated that second-order closure results can be used to estimate the effective turbulent diffusivities for stratospheric flow calculations. Dr. Pao discussed the possibility of using complete numerical solutions of the unsteady Navier-Stokes equations to predict the character of a turbulent flow. The approach would be to obtain Fourier transforms of the flow equations and integrate the results as a function of time. The result for the velocity field could then be obtained at any time by inverting the transform.

Simplified atmospheric models, appropriate for sorting out important chemical reaction sets, were reviewed. The models are basically one- and two-dimensional unsteady kinetics descriptions with parameterized transport/diffusion.

In addition, requirements for and availability of atmospheric data needed in the development and corroboration of atmospheric models were discussed.

Among the conclusions of the Panel were the following (primarily the opinions of Dr. Taylor and Dr. Underwood):

- Simplified, approximate methods can be used to estimate high-altitude-aircraft wake dimensions and species concentrations.
- Two- or three-dimensional, time-dependent turbulent-flow models are needed to treat the dispersion and transport region. The models should include provision for non-equilibrium chemistry. It should be determined if radiation is important on this scale.

- Numerical methods appear adequate for modeling of the wake and dispersion and transport regions.
- Turbulent flow models should consider the effects of turbulence upon reaction rates. Second-order closure and data can be utilized to bound the magnitudes of eddy viscosity. Application of second-order closure except to estimate effects of inhomogeneous mixing upon chemical reaction rates presently appears impractical.
- Presently available global circulation models require major refinements and extensions to be applicable to the CIAP problem.
- It is unlikely that a three-dimensional, global-circulation model with fully interactive chemistry and fluid dynamics can be developed by the 1974 CIAP deadline.
- The suitability of various simplified models for treating global-scale atmospheric motions and physics requires further investigation.
- Attention must be devoted to development of a methodology for coupling models of the wake region, the dispersion and transport region, and the global circulation in order to examine effects of high-altitude aircraft operations.
- Studies are needed to specify accuracy requirements for various atmospheric models applicable to the CIAP and to compare model capabilities with required accuracies.
- Continual and expanded interaction between fluid-dynamic and chemical modelers is essential. Furthermore, close coordination is needed between CIAP atmospheric modeling investigators and other CIAP participants (e.g., those making engine emissions measurements, those projecting 1985-1990 high-altitude aircraft routes, those making atmospheric measurements, and those performing laboratory chemistry measurements).

The remainder of the Fluid Dynamics Panel Report is a compilation of descriptions by Panel members of their ongoing and planned activities pertinent

to the CIAP objectives. Descriptions are grouped generally into the following categories: (a) aircraft wake modeling, (b) dispersion and transport modeling, (c) global-scale circulation modeling, (d) simplified atmospheric models, (e) turbulence modeling, (f) atmospheric data, and (g) model sensitivity studies and intercomparisons.

AIRCRAFT WAKE MODELING

High-Altitude Aircraft Wake Dynamics

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The objective of this program is to develop a mathematical model of aircraft wakes to calculate exhaust species concentrations up to the point where the atmospheric circulation becomes dominant. Input to the model will be emission data, and aircraft and flight parameters. The output will describe the final wake size and shape, and species concentrations in the wake.

The approach is dictated entirely by the needs of the CIAP program, which includes a tight time schedule, parametric rather than detailed calculations, and relatively wide error tolerance in view of basic uncertainties in several portions of the program. As a consequence, the strategy is to develop simple models for the chemistry and fluid dynamics, resorting to complicated, coupled modeling only as dictated by proven necessity. Preliminary studies indicate that uncoupled models for the chemical and dynamical aspects of the wake will be sufficient.

The program involves two main efforts, corresponding to the dynamical and chemical models.

The dynamical model will include interactions between the exhaust gases and the trailing vortex pair generated by the wings, and buoyancy effects due to the high-temperature exhaust and atmospheric stability. The theoretical model will be guided and adjusted according to experimental measurements of aircraft wakes obtained by photographic studies of tropospheric and stratospheric wakes. The chemical model is discussed separately.

The combined chemical and dynamical model is scheduled to be completed in CY 1972, with further adjustments and update contemplated in CY 1973.

DISPERSION AND TRANSPORT MODELING

Subgrid Diffusion

Hugh W. Ellsaesser
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Livermore, California

Between exhaust wake stabilization (approximately two minutes) and the time the wake reaches grid dimensions of a dynamic model (approximately 500 km or 50 hours) there will exist a cylinder of highly concentrated exhaust. To study possible photochemical reactions during this period will require an estimate of concentration as a function of time.

Modifications of the 2BPUFF code developed by Todd Crawford at LLL can be used to look at this problem. This code was developed to predict the concentration of radioactivity as a function of time in clouds resulting from nuclear cratering shots and has been rather extensively tested for this purpose.

The code predicts the diffusive growth of a cylindrical cloud aligned along the vertical. Horizontally isotropic diffusion is based on the Kolmogorov-Obukhov similarity theory of turbulence, in which the eddy diffusion coefficient is a function of the dissipation parameter (epsilon) and the elapsed time. Vertical diffusion is based on K-theory, in which the diffusion coefficient is a function of height and time.

Model input includes epsilon and the vertical profile of eddy diffusivity as a function of time and the dimensions and mass in the initial cloud of the diffusing species. Adaptation to a continuous source can be accomplished by superposition of sequential puffs, as one possibility. Another possibility is to remove cyclindrical symmetry assumptions from 2BPUFF and solve in the Y-Z

plane to study the interaction of several wakes from planes flying normal to the Y-Z plane.

Both the model and the results of comparisons with observations have been documented in internal LLL reports.

GLOBAL-SCALE CIRCULATION MODELING

LLL Zonal General Circulation Model

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Livermore, California

The LLL Zonal Atmospheric Model (ZAM) is a two-dimensional (latitude and vertical) general circulation model, the atmospheric physics of which is derived from Leith's Livermore Atmospheric Model (LAM) by setting all derivatives with respect to longitude equal to zero. In making this two-dimensional assumption, the objective of the model has been changed from attempting to simulate the weather to attempting to simulate the climate—compromising some of the physics, but gaining significantly in reduced computer time (about 45 minutes on the CDC 7600 per year) and in the ability to more easily study a wide variety of climatic simulations. The model, to date, has been used to study several ice age hypotheses.

Specifically, the model uses a 5° latitude grid. There are six vertical levels in a pressure coordinate system which at present reaches only to 100 mb and so does not adequately represent the stratosphere. The primitive (Navier-Stokes) equations are used for the prognostic variables u, v, T, P_s, and water vapor content. In order to simulate the effective Rossby wave (or non-zonal) transport of energy, momentum and water vapor, an eddy diffusion term is introduced in addition to that used to simulate sub-grid scale transport. This mechanism is now being improved to be dependent on internal model dynamics. Cloud cover is a dynamic function of relative humidity and is coupled to both the long-wave and direct and diffuse short-wave (solar) radiation calculations. Precipitation results from both supersaturation and a small-scale convective

prescription. Ozone and carbon dioxide absorption have been only very crudely included, radiation transport being mainly affected by water vapor and cloudiness with a spectrally averaged transmissivity. Daily and annual variation of solar radiation is permitted.

The surface of the earth is presently treated by setting each 5° band to be either ocean, ocean covered with sea ice, or one of two types of land which may be (or become) snow and/or ice covered. From a thermodynamic point of view, the ocean is treated as a single layer, the thickness of which has typically been chosen as the thermocline depth, and an energy balance (solar, long wave, evaporation, sensible) is maintained for the layer. Meridional ocean heat transport has been prescribed to agree with observation and is not dynamically driven by the atmosphere. The ocean may form a sea-ice cover with open leads (treated as a distinct surface type thermodynamically) if the temperature reaches the freezing point. Both sea-ice and the land, possibly snow covered, are divided into a maximum of ten layers for which the one-dimensional energy transport equation is solved, with the atmospheric fluxes applied to the top layer. Snow may melt, there is a very crude runoff prescription, and albedo is changed depending on surface type and condition. A restricted method of introducing mountains exists in order to treat such massifs as the Antarctic continent. At present, the model is being changed to treat a "pointillist" surface; that is, the surface types are proportionally represented at each latitude without establishing a longitudinal grid, each type having its own vertical flux (radiative and convective) pattern, but the individual characteristics being averaged over surface type in the calculation of latitudinal transports.

As to the general utility of the model, both advantages and disadvantages exist. With respect to the latter: (1) the model has not been well verified nor have many of the atmospheric mechanisms been calibrated recently, yet the model does give reasonable meridional sections and a recent comparison with Mintz' three-dimensional model and real data increases confidence that the model can be useful if used carefully; (2) the eddy diffusion prescription is at present very

crude and it is not clear that relatively simple formulations can be developed which will adequately represent stratospheric eddy transport; and (3) the present model has no stratosphere (stratospheric layers are to be included but, as indicated above, the degree of parameterization needed is not yet clear; however, it will probably be much less than a simple diffusion approximation will require). Advantages of the model include ability to run extended periods at relatively low cost, thus permitting a degree of initiative not open for three-dimensional models. Thus, comparison runs (perturbed versus control) are quite easily accomplished, a tool which can be used even if discrepancies exist in comparing the computed climate with the observed. That is, comparison of model runs can often act as a first-order filter of ignorance and allow a determination of the sign of the effect. Also, the relative simplicity of the model aids in tracing causality and thus leads to improved understanding.

During the next two years, we plan to improve ZAM in two different ways. The first is to refine and improve the mechanisms which ZAM now treats, including, for example, surface interaction, the eddy diffusion prescription, and the cloud prescription. The second type of improvement is to extend the model to treat factors important to CIAP objectives, including adding several layers in the stratosphere, refining the radiation budget, and treating several constituents (an appropriate reduced set) and photochemistry. Based on additional efforts being undertaken at LLL in photochemical kinetics, spectrally dependent radiation studies, and a plume-diffusion treatment of the aircraft wake, we expect to undertake extensive numerical experiments and parameter studies intended to define the bounds of potential climatic consequences of SST operations as compared to the bounds of natural climatic variations.

With the model undergoing extensive revision, only the source deck of the early version is readily available. Proposed experiments for the model are welcome, although availability of access and support is subject to certain

AEC restrictions and to present plans for use. Literature on the model is available through internal reports from M. MacCracken (Lawrence Livermore Laboratory, P. O. Box 808, Livermore, California 94550) on request.

M.I.T. Plans for Global-Scale Circulation Modeling

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The M.I.T. group intends to reassess certain features of the "unperturbed" stratosphere and ozone distribution and to explore the perturbing effect of the projected 1990 SST operation (500 planes) through two closely related tasks, viz, (a) the development of a numerical dynamical model and (b) an investigation of the effect of trace constituents on heating rates.

The dynamical model to be developed will be simpler than the full-scale dynamical models (which typically use about 10^4 grid points). The fact that we are interested primarily in the stratosphere, with a relative absence of small-scale structure, enables a simpler model to be used with a considerable claim to reality. In this model a fixed smallish number (N) of spherical harmonics will be used for the horizontal representation cf the dependent variables. This procedure has been used for the past ten years in dynamical meteorology (Baer and Platzman, 1961) and is currently undergoing considerable development (see, for example, Orszag, 1970, where a method of speeding up the calculation is developed suitable for large N). We expect to use 42 spherical harmonics (N = 42) corresponding to six latitudinal wave modes for each of seven longitudinal wave numbers, S. Values for S of -6, -4, -2, 0, +2, +4, +6 will recognize the quasi-stationary "long waves" with $S = \pm 2$ and the baroclinically unstable shorter waves with $S = \pm 4$ and ± 6. These will interact with one another and the

main flow $S = 0$. In the vertical direction we plan to represent the dependent variables on approximately 20 pressure surfaces located between the ground and about 60 or 70 km. A doctoral thesis completed in the Department of Meteorology recently by K. Trenberth (1972) successfully used a model of this type to study stratospheric warmings. His model reproduced realistic stratospheric warmings in late winter. Based on his numerical experience, we estimate about 12 hours of computer time required on the M.I.T. IBM 360 for a one-year integration of our model (which will include certain chemical reactions). The model will be based upon the quasi-geostrophic system (Phillips, 1963). Although Trenberth neglected the chemistry, Clark (1970) had previously used a similar model (with somewhat fewer spherical harmonics and coarser vertical resolution) which did include the simple Chapman scheme of oxygen photochemistry. Clark was able to reproduce numerically the observed tendency for the large-scale motion field to shift the maximum of vertically integrated ozone poleward from the low-altitude source region. In such a model the chemistry interacts with the dynamics via the heating rate Q, which will depend not only on infrared radiation and a parameterization of small-scale convection in the troposphere, but also the absorption of radiation by the variable amount of ozone. NO_x will be added to the reaction scheme in our model and initially we shall operate with a prescribed distribution of NO_x. More interesting, however, will be later computations in which odd nitrogen is also forecast via a species conservation equation.

As a check on the parameterization of heating rates in the numerical model, and as information valuable in its own right, we shall perform more accurate heating rate computations using a computer program already available at M.I.T. (Dopplick, 1970). Dopplick's program will, however, be extended to include altitudes above 30 km and the effects of aerosols on the heating rates.

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NCAR Plans for Global-Scale Circulation Modeling

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The tropospheric general circulation model (GCM) developed at NCAR (see, for instance, Kasahara and Washington, 1971) has recently been modified to include 12 layers with the top layer extended to 36 km. The expanded model required incorporation of additional physical processes operating in the stratosphere. A notable new physical factor is the calculation of solar and terrestrial radiation within the stratosphere. Although the solar energy absorbed in the stratosphere is relatively small, its heating effects are important since the density and therefore the specific heat capacity of the stratosphere are also small. In the first version of the stratospheric model, an observed climatological distribution (as a function of latitude and height) of ozone is used to avoid complication due to photochemical reactions. Also, constant mixing ratios for water vapor and carbon dioxide in the stratosphere are assumed. These distributions are used for both the solar and terrestrial radiation calculations. Cloud distributions are computed at 3- and 9-km levels as in the tropospheric model. It is expected that as this study progresses the resolution in the vertical will be changed to suit various experiments and that the physics will be modified and improved.

The photochemical model to be used in the GCM will include oxygen-hydrogen and oxygen-nitrogen interactions with assumed fixed water vapor and NO_x mixing ratios in the stratosphere. At a later stage, it is proposed that important hydrogen-nitrogen interactions be included and that perturbations to the water vapor and NO_x distributions be added to simulate possible SST effects.

In the numerical computations, the stratospheric circulation would still be generated by radiative heating functions derived from fixed (observed) ozone, carbon dioxide and water distributions. The resulting three-dimensional stratospheric circulation would be used to transport ozone until an approximate near equilibrium is reached (presumably after about 60 days), with the observed latitudinal ozone cross section taken as the initial distribution. The ozone equation would add the appropriate photochemical sources and sinks to the total transport terms.

The model will be based on current values of the photochemical parameters. However, the reaction rates for some of the reactions (e.g., $O(^1D) + H_2O$; $O_3 + OH$; $O_3 + HO_2$) are not very well known. New data will be incorporated into the model as they become available from the experimental studies presently being conducted in various laboratories. Also, tests will be made of the sensitivity of the results to the range of uncertainty in the various reaction rates.

A numerical experiment will be carried out for both hemispheres to approximate, for instance, mean January and July conditions. The finally derived ozone distribution will be compared with the observed distribution. The validity of the photochemical-circulation model can thus be checked and it should be possible to assess the importance of stratospheric contaminants in disturbing the "normal" ozone distribution.

Information on the exhaust products of contemplated SST flight programs and the long-term average concentration and three-dimensional pattern of distribution of these contaminants will be used to perturb the "normal" ozone distributions (through photochemistry). The resulting stratospheric and total ozone distributions will be used to evaluate variations, if any, in the atmospheric ultraviolet transmission characteristics.

Throughout the study, close cooperation will be maintained between project scientists in the Astro-Geophysics Department at the University of Colorado and NCAR scientists.

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GLOBAL-SCALE CIRCULATION MODELING

Tentative GFDL SST-Related Research Plans

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Tentative GFDL plans for SST-related research are summarized below. Parts related to the general climate change problem will be done. Those parts specifically related to the SST problem may or may not be executed in the near future. Note that no step can be done until the previous steps have been accomplished:

- Three-dimensional tracer experiments using well-known tracers (such as nuclear tests)
- Three-dimensional tracer experiments for a simulated SST-type source with a ground-level sink (normalized dispersion for H_2O, NO_x, and particulates)[1/]
- Once our ozone photochemical scheme has been thoroughly checked out in a simpler framework, we plan to do a non-interactive ozone photochemistry test using model tapes (may not be done until newest model is converted to the Texas Instruments ASC machine)
- General circulation experiment using "free" model-determined ozone in the radiative transfer code
- Fully interactive (by SST sources of H_2O, NO_x, and possibly particulates) general circulation climate experiment.[1/]

[1/] Support dependent.

UCLA Plans for Global-Scale Circulation Modeling

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The current UCLA numerical general circulation model covers only the global troposphere. The characteristics of this model have been described by A. Arakawa (Design of the UCLA General Circulation Model, Department of Meteorology, UCLA, 1972, 122 pp.) A detailed documentation of the two-level version of the model has been given by W. L. Gates et al (A Documentation of the Mintz-Arakawa Two-Level Atmospheric General Circulation Model, RAND Report R-877-ARPA, 1971). This model has been highly successful in simulating the observed planetary-scale circulations of the troposphere.

We are now in the processes of extending the upper boundary of the model to the stratopause. This will involve not only increasing the number of levels, but also possible reformulations of the finite-difference scheme and the upper boundary condition in order to obtain more accurate simulation of the vertical propagation of wave energy. The radiative transfer calculations of the model will be expanded to include the transfer by stratospheric ozone. We shall also attempt to parameterize the photochemical sources and sinks of ozone, including the influences of NO_x and possibly of water vapor compounds.

A critical test of the model will be its ability to simulate the following thermo-hydrodynamic phenomena of the stratosphere:

- The vertical and horizontal thermal structure of the stratosphere, including the tropopause and its breaks

- The seasonal variations of the stratospheric temperature and motion fields, including the change from summer axisymmetric easterlies to winter westerlies with predominant wave numbers one and two
- The transient sudden winter warmings and accompanying breakdowns (and restorations) of the stratospheric polar night jet, and the final abrupt springtime transition to summer axisymmetric easterly flow
- The simulation of the stratospheric equatorial waves and the biennial oscillation of the equatorial stratosphere.

When we are able to successfully simulate the above properties of the natural stratosphere, we will use the model to simulate the atmosphere when it is perturbed by engine emissions of projected supersonic jet aircraft. Given the space-time source distribution of the jet emissions, we will study the horizontal and vertical transports and photochemical evolution of the aircraft pollutants (a) under the assumption of no influence of the pollutants on the circulation, and (b) taking into account the photochemical and radiative feedback of the pollutants upon the circulation.

LLL Global Box Model

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As part of its work under CIAP, LLL plans to develop what is being called a global box model. The underlying motivations of this effort are twofold: (a) it is a relatively simple global transport model which can treat chemical kinetics and will be an important tool for narrowing the range of possibilities which will have to be treated in the more sophisticated models; and (b) we believe that the data on the stratospheric concentrations of radionuclides provide the most definitive clue to the transport of tracers in the upper atmosphere and should therefore be used to the fullest extent possible by CIAP.

The concept of a box model approach (which can be defined as an Eulerian grid model in which the transport terms are prescribed as diffusion coefficients, residence times, or effective transport velocities—all of which serve to average over the effects of the actual mean and eddy wind components—such that the net result of using these values yields the observed tracer distribution) has a long history. Some of the present proponents are Friend (NYU), Krey (HASL), and Libby (UCLA). This almost heuristic approach has not been intensively pursued recently, we believe in great part because many conclude that the final answer must come from detailed general circulation models based on the primitive equations. Yet it is clear that this latter will not be achieved within the CIAP timetable. Therefore, we at LLL feel the box model approach is worthy of further attention, even though the assumption must be made that the circulation and chemical-radiative effects can be decoupled.

Actually, many of the kineticists have been using a one-box model of the stratosphere to study a static atmosphere, throughout which they mix the

SST effluent. The radionuclide data available from aircraft and balloon sampling justifies a significantly more refined grid, perhaps even including a longitudinal structure in order to evaluate build-ups over or downwind of flight routes. The details of model structure have not yet been defined, in part because the time and space scales of inert tracer transport will dictate the need for inclusion of photochemical and radiative effects, which in turn may influence the grid structure. In summary, however, the intent is to utilize the observed concentration data (rather than the observed circulation patterns) as a means of understanding the effective transport of tracers in the stratosphere, giving, special care to the interpretation of results in view of the assumptions made.

Two-Dimensional Time-Dependent Photochemical Model Including Effects of Vertical Mixing

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We have computerized a one-dimensional, time-dependent model that includes the effect of vertical mixing by eddy diffusion. The program can calculate vertical distributions of densities of many minor constituents by solving their continuity equations simultaneously. A special iteration technique has been used for obtaining conservation of particles for solving many nonlinear finite-difference equations for n_i's simultaneously by the implicit method (Shimazaki, 1972). The program was originally developed for the study of the mesosphere and lower thermosphere (Shimazaki and Laird, 1970 and 1972), but it was applied to the stratosphere (Ashby, Shimazaki and Weinman, 1972).

We plan to extend our program to a two- or three-dimensional model for the stratosphere. Considering first a steady state condition on a 360° meridional plane, we will solve the two-dimensional continuity equations, represented by spherical coordinates, for day- and night-side zonal-averaged densities. Our domain includes the summer and winter condition as well as the night and day condition. The side boundary condition will be given by the requirement of cyclic variations for density distributions. We will use information for temperature and wind structure from either observations, or results of global circulation models. The problem will eventually be solved as a time-dependent problem; the model then includes diurnal or longitudinal variations and could be a kind of three-dimensional model.

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SIMPLIFIED ATMOSPHERIC MODELS

Ames Model of Stratospheric/Mesospheric Photochemistry

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The Ames model is very similar to that described by Dr. Shimazaki, i.e., it obtains solutions to a one-dimensional (vertical) set of coupled transport equations that include the photochemistry as well as transport which is parameterized by means of eddy diffusion coefficients. It extends from an altitude of 10 km to 90 km and contains about 28 constituents, which include the oxygen-hydrogen-nitrogen-carbon system. Sulphur compounds (which produce the aerosol layer at 20 km) will be introduced at a later date. Unlike the Shimazaki model, we do not try to prevent apparent loss of hydrogen through computer error; this simplification is justified because we limit our upper boundary to 90 km where the dissociation of water vapor is just beginning to become appreciable. We also neglect the effect of turbulence on the reaction rates. The boundary conditions have been selected in a somewhat arbitrary manner, some constituents being chosen to be in photochemical equilibrium at the boundaries, others have some flux condition and others have a fixed concentration at the boundary. For example, nitric oxide has a downward flux of 10^8 cm^{-2} sec^{-1} at 90 km and is in mixing equilibrium at 10 km. These boundary conditions will be varied within "reasonable" limits in order to determine the effects. In many cases, the choice of boundary conditions is not very important except close to the boundary.

The functions of this model are several: (a) in a rather coarse way, evaluate the effects of an SST fleet and space shuttle on the minor constituents in the stratosphere and mesosphere; (b) obtain a better understanding of the photochemistry of the unperturbed atmosphere; (c) carry out a sensitivity analysis of

the various reactions in order to simplify the photochemistry used in more detailed models; and (d) identify those important reactions whose rate coefficients are not yet adequately known.

At the present time, the complete model is just beginning to work. The applications enumerated above will probably take about 6 to 8 months to complete. As may be expected, our results are not significantly different from those of Dr. Shimazaki. Like Dr. Shimazaki, we are currently working on a two-dimensional model with parameterized transport.

Turbulent Diffusion and Chemistry Modeling Using Second-Order Closure

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Among the problems considered by CIAP, the following are particularly well-addressed by the recently developed techniques for modeling turbulent diffusion and chemistry using Donaldson's invariant version of second-order closure:

- The lateral and vertical spread of exhaust emissions up to the time the plume dimensions are of the order of the grid scale used in global circulation models
- Estimation of the lateral and vertical eddy diffusivities which should be employed in the global circulation models so as to assure adequate representation of diffusive mixing
- Investigation of the effects of inhomogeneous mixing of reacting materials, particularly for relatively fast reactions of importance in photochemical chains.

Second-order closure, while demanding of computers, offers valuable insights into the physical processes which are operative in diffusion and chemistry of exhaust materials and ambient constituents since second-order correlations are specifically retained. In the diffusion calculations, for example, only the mean wind speed and temperature profiles and the initial pollution concentration distribution need be specified; no a priori assumptions with regard to subsequent distribution functions or effective diffusivities are involved. This model may be used either directly or as a basis for improved estimates of the effective diffusivities which should be used in first-order closure models.

The fact that inhomogeneous mixing has a significant effect on chemical reaction rates has been known for some time. The work of O'Brien and Donaldson and Hilst has only recently provided a theoretical basis for analyzing the magnitude of this effect and its causes. These analyses have shown that chemical reaction rates may be suppressed by as much as two to three orders of magnitude when the characteristic reaction time is fast with respect to the characteristic molecular diffusion rate. A large family of reactions of importance exhibit this property.

The mathematical models developed by O'Brien and Donaldson and Hilst provide an initial method for estimating the magnitude and duration of this effect in the high atmosphere. The invariant model, when appropriately developed for arbitrary concentration distributions, is particularly appropriate since the chemistry can be interactive with the diffusion processes.

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TURBULENCE MODELING

Numerical Simulation of Turbulence and Turbulent Mixing with Chemical Reaction

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Orszag and Patterson (1972) have simulated homogeneous turbulence on a CDC 6600 computer at NCAR, utilizing the spectral method developed by Orszag (1969, 1970, 1971). Their computational results agree well with wind tunnel measurements of Stewart and Townsend (1951). This work is indeed very exciting and has opened up a new chapter in turbulence research.

Flow Research, Inc., in cooperation with S. Orszag, is now extending this work to turbulent shear flow with simple geometries where the turbulence is non-homogeneous. This work also includes the effect of density stratification. Results from these computations will give the statistical details of these simple turbulent shear flows and enable us to examine a number of turbulence closure schemes that are used in numerical models.

This work can be extended readily to include turbulent mixing with chemical reaction and enable us to examine critically the effect of turbulent mixing on chemical reactions.

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ATMOSPHERIC DATA

Tasks of the M.I.T. Planetary Circulation Project Related to the Needs of CIAP

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The main tasks of the Planetary Circulation Project at M.I.T. are to gather, edit, and analyze atmospheric data in order to understand the dynamics of the general circulation of the earth's atmosphere.

Presently, the MIT General Circulation Data Library consists of five years of radiosonde data covering the years 1958 through 1963. The data has been collected from stations located over the entire globe and analyzed in detail. Approximately 700 of the source radiosonde stations exist in the Northern Hemisphere and about 120 in the Southern Hemisphere. Wind speed, wind direction, temperature, and relative humidity are the data collected. Typically, more than 20 levels are available for each observation up to a maximum of 7 mb in altitude. One observation per day normally is available. The data have been carefully edited and exist in two forms: (a) map format—every station is recorded sequentially for each day on tapes; (b) station format—one station is recorded on tape over five years; then the next station over five years, etc. Relative humidity data is reliably available up to about 300 mb. In the stratosphere, wind and temperature are available for approximately 40 to 50% of the potential observations over five years at 20-km altitude, and are available for approximately 15 to 18% of the potential observations at 30 km.

The further intentions of M.I.T. and ERT are to concentrate heavily on developing techniques to utilize data sources other than radiosonde, in particular, satellite data for the higher levels.

Associated with the data sets is a sophisticated software package which allows hemispheric arrays of data to be easily manipulated. This hyper-language allows two- and three-dimensional analyses to be performed with relative ease and with a minimum of computational error. Vector operators are part of the available software.

The data collection and associated software are available for general use

In the next two years, the intention of this group is to collaborate with the Geophysical Fluid Dynamics Laboratory of NOAA to extend the data base from the present end point in 1963 through 1973.

ATMOSPHERIC DATA

M.I.T. Meteorology Department Data Analysis Plans

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In addition to our modeling activities, we intend to analyze a complete and consistent set of five years of observational data in the stratosphere. The data to be analyzed comprise the field of geopotential height and temperature at the 100, 50, 30 and 10 mb pressure levels for the years 1963-1967 for latitudes poleward of 20°N. The data has been obtained from objective analysis of radiosonde observations. The analysis will give information necessary to specify the dynamic climatology of the region in terms of the temperature and motion fields and the components of the mass, momentum, and energy budgets. This data analysis will provide a basis against which both the numerical model and possible SST-induced perturbations may be compared.

MODEL SENSITIVITY STUDIES AND INTERCOMPARISONS

RAND Plans for CIAP-Related Modeling

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RAND has proposed two tasks related to the topics discussed during the CIAP Computational Modeling Workshop. The first task is a study of the validation of numerical models to simulate climatic change. The second task is a detailed analysis of the effects of particulate matter in the atmosphere on global climate.

a. The Validation and Sensitivity of Global Circulation Models— This task will attempt to answer the question whether global circulation models actually simulate a real atmosphere and, if not, under what conditions their predictions of the effects of stratospheric pollution are valid. This study will make available to workers in global simulation a collection of statistical tests and comparison techniques by which they can assess the performance of their simulation experiments. This work falls into three categories:

   1. Formulation of a climatic data base and comparisons of simulations with real climate. For example, could the simulated global-averaged June be a sample from the distribution of all global-average Junes?
   2. The assessment of model sensitivity to small changes in its boundary and initial conditions. Did we, in fact, actually modify the climate, or is what we detect due only to the model's lack of predictability?
   3. Inter-model comparisons.

b. Investigations of the Climatic Effect of Particulate Matter in the Atmosphere—The main thrust of this program will be to begin a systematic investigation of the effects of various amounts and distributions of particulate matter on the general circulation of the atmosphere. This work will proceed in four steps:

1. Estimate the number density and size distribution of particulate matter as a function of latitude, longitude and height
2. Estimate scattering and absorption of solar radiation by the particulate cloud
3. From the calculations above estimate new parameters of the solar radiation reaching the surface of the earth
4. The parameters obtained above will be inserted into the global circulation model in use at RAND to determine the extent of changes in the circulation and other meteorological parameters. The procedures developed in Task a will be used to determine the validity of the results.

III. REPORT OF THE CHEMICAL MODELING PANEL

H. Harrison, Chairman

OVERVIEW

The Chemical Modeling Panel addressed itself to the following tasks:

- Assemble an inventory of plausibly important chemical reactions which may affect the stratosphere.
- Organize these into an appropriate hierarchy of importance, and comment on the approximate precisions with which rate constants are known.
- Examine possible influences of chemistry in the immediate wake of high-altitude aircraft.
- Examine possible rate effects due to concentration fluctuations in the stratosphere as a whole.
- Examine possible chemical influences of aerosols.
- Comment on appropriate numerical methods for chemical modeling.
- Discuss and recommend priorities for measurements of the chemical state of the stratosphere.

- Suggest ways by which chemical and fluid dynamic models may interact with measurement programs.

The remainder of the Chemical Modeling Panel Report is divided into seven sections, one dealing with each of the above-mentioned tasks. Each section reports the findings of the Panel pertinent to the subject of the section.

REPORT OF THE REACTION RATE SUB-PANEL

M. H. Bortner - Chairman

G. Brasseur
R. Gelinas
T. Hard
F. Hudson
H. S. Johnston
R. H. Kummler
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and others

Introduction

The influence of trace species upon the energy balance of the earth will ultimately depend upon the chemical balance of these trace species, globally integrated. The single most important parameter which is needed to describe the rate processes which define that balance is the reaction rate constant. The complex set of governing equations to be discussed herein requires the knowledge of at best a dozen rate constants and at worst several hundred, depending upon the altitude range, the latitude and solar cycle range, and the degree of perturbation which the kinetics are expected to describe. We have endeavored herein to outline the kinetics primarily associated with the O_3 balance. We have considered to that end:

- A list of reaction processes partially ordered as to importance
- Recommendations for additional laboratory work, based on our present knowledge of the rate constants for those processes
- The need for a newsletter among interested parties, as a rapid route for the distribution of new rate constant information

- Suggestions of novel reaction schemes which need further work
- A recommendation for updated atmospheric composition information needed to assess the relative importance of various reactions
- Preliminary information on the uncertainties of the rate constants necessary to perform an adequate sensitivity analysis.

A critically important responsibility of the CIAP program will be to provide sensitivity studies which can place error bars or confidence limits on the final results of both the coupled overall solutions and the individual modules of the program. In particular, parameters will be varied to suggest effects upon the solutions. It is important that these variations be confined to physically plausible regions. It is for this reason that the reaction-rate sub-panel has provided confidence limits for the rate-constant estimates wherever possible.

It is realized that species other than O_3, particularly H_2O and CO_2, can affect the atmospheric thermal balance. A secondary study to cover factors influencing these seems advisable.

Reactions And Rate Constants

Table 3.1 presents a list of reactions that are believed to control the ozone concentrations both in the natural atmosphere and in the atmosphere disturbed by an SST. Although an attempt has been made to make this list as complete as possible—or as complete as necessary to permit the calculation of ozone to the accuracy desired—there may be omissions (due to the short time available) which were not intended. In addition to the reactions, uncertainties in our knowledge of the rate constants are indicated.

TABLE 3.1. REACTIONS AND CONFIDENCE LIMITS FOR THEIR RATE CONSTANTS

Note: This table is a corrected version of Table 2 (pp. 4 and 5) of Appendix B.

Reaction No.	Reaction	Confidence Statement
	I. Basic Chapman Mechanism	
1	$O_2 + h\nu \rightarrow O + O$	$\sigma \pm 20\%$
2	$O + O_3 \rightarrow O_2 + O_2$	k well known
3	$O + O_2 + M \rightarrow O_3 + M$	k well known
4	$O_3 + h\nu \rightarrow O(^1D) + O_2$	$\sigma \pm 10\%$
	II. Destruction of Odd-Oxygen by NO_x	
5	$NO + O_3 \rightarrow NO_2 + O_2$	$k \pm 20\%$
6	$NO_2 + h\nu \rightarrow NO + O$	σ, J $\pm 20\%$
7	$NO_2 + O \rightarrow NO + O_2$	k + 20, −50%
	III. Removal of NO_x	
8	$OH + NO_2 \xrightarrow{M} HNO_3$	k factor of 2, function of pressure
9	$OH + HNO_3 \rightarrow H_2O + NO_3$	k factor of 2
10	$HNO_3 + h\nu \rightarrow H + NO_3$	σ factor of 100
11	$\rightarrow OH + NO_2$	
	IV. NO Production	
12	$N_2O + O(^1D) \rightarrow NO + NO$	k factor of 2
13	$\rightarrow N_2 + O_2$	k factor of 2
14	$N_2O + h\nu \xrightarrow{\sigma_I} N_2 + O(^1D)$	$\sigma_I \pm 20\%$
	IV.A. Hypothetical Reaction Affecting NO	
15	$N_2O + h\nu \xrightarrow{\sigma_{II}} N + NO$	Important if $\sigma_{II} = .01$ $\sigma_I(N_2O + h\nu \rightarrow N_2 + O)$
16	$N_2(A) + O \rightarrow NO + N$	
17	$O(^1D) + M \rightarrow O(^3P)' + M$	$O(^3P)'$ indicates hot $O(^3P)$
18	$M + O(^3P)' + N_2 \rightarrow N_2O^* + M$	N_2O^* indicates excited N_2O
19	$N_2O^* \rightarrow NO + N$	
20	$\rightarrow N_2 + O$	
21	$\rightarrow N_2O$	
22	$O(^1D) + N_2 \rightarrow N_2O^*$	Slow
	V. Higher Oxides of Nitrogen	
23	$NO_2 + O_3 \rightarrow NO_3 + O_2$	k factor of 10^4 uncertainty at stratospheric T depending on activation energy = 7 ± 2 kcal.
24	$NO_3 + h\nu \rightarrow NO + O_2$	σ, J $\pm 20\%$ at peak (6200A) ϕ factor of 10
25	$NO_3 + h\nu \rightarrow NO_2 + O$	σ small at peak, possibly important at lower wavelength

σ – Photochemical absorption cross section
k – Reaction rate constant

TABLE 3.1 (Cont)

	V. Higher Oxides of Nitrogen (Cont.)	
26	$NO_2 + NO_3 \rightarrow N_2O_5{}^*$	k ± 20%
27	$N_2O_5{}^* \rightarrow NO_2 + NO_3$	
28	$\rightarrow N_2O_5$	
29	$N_2O_5 + h\nu \rightarrow$	Products unknown, being worked on
30	$N_2O_5 + O \rightarrow$	
31	$N_2O_5 + OH \rightarrow$	
32	$N_2O_5 + HO_2 \rightarrow$	
33	$N_2O_5 + H_2O \rightarrow$	Slow in gas phase
	VI. $O(^1{}_D)$ and O_3	
34	$O(^1{}_D) + O_3 \rightarrow O_2 + O_2$	
35	$O(^1{}_D) + M \rightarrow O(^3{}_p) + M$	
	VII. Reactions Affecting OH, HO_2	
36	$OH + O \rightarrow H + O_2$	k factor of 2
37	$H + O_2 + M \rightarrow HO_2 + M$	k factor of 2; probably only important H reaction
38	$HO_2 + O \rightarrow OH + O_2$	k not known
39	$OH + O_3 \rightarrow HO_2 + O_2$	k factor of 10^3
40	$OH^\dagger + O_3 \rightarrow H + O_2 + O_2$	k depends on vibrational T; † indicates vibrational excitation
41	$OH + CO \rightarrow CO_2 + H$	k factor of 2
42	$O(^1{}_D) + H_2O \rightarrow OH + OH$	k factor of 2
43	$O(^1{}_D) + CH_4 \rightarrow CH_3 + OH$	k factor of 2
44	$O(^1{}_D) + H_2 \rightarrow OH + H$	k factor of 2
45	$OH + HO_2 \rightarrow H_2O + O_2$	k of 2×10^{-10} seems high
46	$OH + OH \rightarrow H_2O + O$	k factor of 2
47	$H + O_3 \rightarrow OH^\dagger + O_2$	k factor of 2
48	$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$	k factor of 2
49	$H_2O_2 + h\nu \rightarrow OH + OH$	σ factor of 2
50	$H_2O_2 + O \rightarrow HO_2 + OH$	k factor of 2
51	$H_2O_2 + OH \rightarrow H_2O + HO_2$	k factor of 2
52	$HO_2 + NO \rightarrow OH + NO_2$	k probably high
53	$HO_2 + CO \rightarrow OH + CO_2$	k low, factor of 10
54	$OH + CH_4 \rightarrow H_2O + CH_3$	k factor of 2
55	$OH + H_2 \rightarrow H_2O + H$	k factor of 2
56	$OH + h\nu \rightarrow O + H$	σ very uncertain
57	$HO_2 + h\nu \rightarrow H + O_2$	σ very uncertain
58	$CH_3 + O_2 + M \rightarrow CH_3O_2 + M$	
59	$CH_3O_2 + NO \rightarrow CH_3O + NO_2$	
60	$CH_3O_2 + CH_3O_2 \rightarrow CH_3O + CH_3O + O_2$	
61	$CH_3O + O_2 \rightarrow CH_2O + HO_2$	
62	$CH_2O + OH \rightarrow HCO + H_2O$	k factor of 2
63	$HCO + O_2 \rightarrow CO + HO_2$	k high; uncertain but sufficient

The first group of reactions given constitutes the basic Chapman mechanism which, in a pure oxygen atmosphere, controls the ozone concentration. Following this group of reactions is given a hierarchy of groups in decreasing order of importance. Group II reactions are those which destroy odd-oxygen species (O_3, O, etc.) and largely determine the NO/NO_2 ratio. Group III introduces removal mechanisms for NO and NO_2, providing conversion to HNO_3, etc. Following this are four groups which control or significantly affect the concentrations of the species in the first three groups. Group IV reactions provide for stratospheric production of NO. Other rather speculative reactions are added in IVa. These are reactions which could be important if they occur and if their rate constants are large enough. The reactions involving NO_3 and N_2O_5 are in Group V while O('D) processes are given in VI. The reactions involving and largely controlling OH and HO_2 are listed in Group VII. The highest accuracy requirements are for rate constants of reactions of Group I and the least accuracy is required for those of the last groups.

It should be emphasized that certain sets of reactions are of importance because they form a cycle. Thus, Group II reactions constitute a cycle which sets up the NO/NO_2 ratio and determines the concentration of NO (and its efficiency to destroy ozone) as a result of side reactions involving NO_2 and O.

It is undoubtedly possible to simplify this system considerably both by the deletion of numerous reactions and by the combining of reactions into "lumped" reactions. An example of one possibility that was suggested is the system:

$$O('D) + CH_4 \rightarrow CH_3 + OH$$

$$CH_3 + O_2 + M \rightarrow CH_3O_2 + M$$

$$CH_3O_2 + CH_2O_2 \rightarrow CH_3O + CH_3O + O_2$$

$$CH_3O + O_2 \rightarrow CH_2O + HO_2$$

$$CH_2O + OH \rightarrow HCO + H_2O$$

$$HCO + O_2 \rightarrow CO + HO_2$$

which could possibly be simplified to

$$O('D) + CH_4 + 3O_2 \rightarrow CO + 2OH + 2HO_2$$

It is, of course, realized that this would require investigation but it is presented as an example of a possible "lumped" reaction. Many of these should be possible, particularly in the lower group of reactions.

The sub-panel recommends that this list of reactions be examined more critically in the near future, and that recommended values of rate constants be given for all of these. Such values should be made available almost immediately and be kept up to date by frequent issuing of a "newsletter" type of publication. The rate constants should be presented in a standard form, e.g.,

$$k = (a \pm \Delta a)\, T^{(b \pm \Delta b)}\, e^{-(c \pm \Delta c)/T}$$

where for practically all reactions one of the temperature terms should be eliminated, by setting $b = 0$ or $c = 0$, since data on the rate constants are not good enough to separate out the two possible dependencies. It is important to include the uncertainties both in the pre-exponential and the exponential terms. Units used should be such as to agree with the NBS publications, specifically those using molecules cm^{-3} for concentration, temperature in K, and activation energy given in K, i.e., as E/R.

It should be noted that certain categories of reactions which do not appear to exist at present would be of first-order importance if found. This, of course, would include any catalytic cycles which reproduce the species destroying ozone. Reactions which do not have this catalytic property are not likely to be important at concentrations of reactants which are reasonably

expected. In addition, reactions destroying odd oxygen* through reaction with O and reactions which either produce or destroy odd nitrogen at rates comparable to the $O^1D + N_2O \rightarrow 2NO$ reaction (1-100 cc^{-1} sec^{-1}) would be very important.

We also note that many reactions of no direct atmospheric interest may be important to measurement techniques of direct interest. These include relaxation and energy transfer reactions which determine the quantum yield of fluorescence measurements, as well as chemiluminescent reactions which would compete or interfere with NO and O_3 measurements using the chemiluminescent techniques.

Ion Chemistry

There appear to be three ways whereby ionic reactions might be significant in the chemistry of the lower stratosphere: (a) direct reactions of ions with ozone; (b) reactions of ions with neutral species that affect the concentration of ozone; and (c) reactions leading to the formation of aerosols.

Each possibility is considered separately below:

- At 20 km, the concentration of any given ionic species is less than 1.5×10^3 cm^{-3}. Ozone concentration is about 3×10^{12} cm^{-3}. To be significant, direct reactions would have to be part of a catalytic cycle involving regeneration of an earlier ionic species. The rate-limiting step in such a cycle would have to be extremely fast, and no such cycles have been proposed.

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* "odd" refers to the atomic parity; thus O, O_3, NO, NO_2, HNO_3 are odd in oxygen or nitrogen, and N_2, O_2, etc., are even.

- Possible ionic mechanisms for the creation or destruction of neutral species, such as the conversion of NO_2 to HNO_3, are limited by ion production and recombination rates relative to neutral mechanisms producing and destroying neutral species. Upper limits to the recombination of ions at 20 km are of the order of 1 cm^{-3} sec^{-1}, and lower limits to the production of NO_x are of the same order. Such ionic mechanisms do not seem likely in the lower stratosphere, but should not be forgotten higher up.

- Ions may serve as nucleation sites for aerosol formation. Within the 1974 CIAP deadline, most useful information on stratospheric aerosol formation will likely be obtained experimentally rather than by chemical modeling.

Stratospheric Composition

Throughout the discussions of the Reaction Rate Sub-Panel, the desirability of ordering priorities was continually expressed, but concrete conclusions were often precluded, more by the dearth of ready knowledge of atmospheric concentrations than by rate constants. While it is recognized that a "final" atmospheric composition appropriate to the CIAP program awaits in most cases a fully interactive chemical-dynamic three-dimensional model, it is also recognized that in many cases large simplification will result from initial ability to discard insignificant processes. Thus, it is important to have available best estimates of the atmospheric composition together with error bounds, so that rates as well as rate constants can be estimated within these error bounds. Such information has been presented by Bortner and Kummler, but as noted by them, it was primarily aimed at the mesosphere and above. Their report is also four years old and should be updated for the CIAP task. Such revision will help in the computation of rates and will provide tentative design goals for experiments aimed at our knowledge of the composition.

This revision should serve as a guide or benchmark for the overall CIAP program. A benchmark atmosphere can be used as a start for many modeling calculations. These will include estimating order-of-magnitude effects of reaction as well as computing changes due to SST effluents. A benchmark model atmosphere should include working values (to be updated periodically) of:

- Temperature, T
- Density, ρ
- Concentrations of minor constituents, N_i
- Optical absorption cross section, $\sigma_a(\lambda, T)$
- Solar flux outside earth's atmosphere, with 1 AU resolution, $\phi_{oo}(\lambda)$
- Solar flux as a function of altitude, $\phi(\lambda, h)$
- Quantum yield for process (ℓ) of interest, $\phi_\ell(\lambda)$. Note that the cross section for a specific process

  $$\ell \text{ is } \sigma_\ell(\lambda) = \sigma_a(\lambda)\phi(\lambda)$$

  This is the quantity that is generally used.
- Photo-dissociation rate per molecule, $J_i(h)$. This first-order rate constant is generally model dependent through the solar flux and concentrations, but ball-park numbers would provide a useful guide.

AIRCRAFT WAKE CHEMISTRY

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Two characteristics of the near-field aircraft flow are recognizably different from those of the rest of the CIAP domain: (a) the exhaust temperatures are much higher than ambient temperatures; and (b) the fluid dynamic motions imparted by passage of the aircraft are distinctly different from the motions normally present in the ambient stratosphere.

Also, to a greater extent than in any other regime, the concentrations of potentially important chemical reactants are higher in the wake than elsewhere.

Because the conceivable stoichiometric changes in the wake cannot appreciably affect the wake energy balance, the wake chemistry can be considered to be uncoupled from the wake fluid dynamics. Thus the wake chemistry starts with the temperature and pressure fields. The chemical analysis then proceeds to consider which engine exhaust products may react in the wake, the mechanisms for these reactions and their progress in the wake regime.

Table 3.2 summarizes preliminary observations on specific chemical reaction systems in the wake regime. The table identifies the reaction system and specifies pertinent conditions in the first two columns. The next four columns list conditions which may limit the significance of a particular reaction system in the wake regime. An X in one of these columns signifies that a preliminary analysis shows the importance of this reaction system to be small in the wake regime.

TABLE 3.2—PRELIMINARY OBSERVATIONS ON CHEMICAL REACTIONS IN THE WAKE REGIME

Reaction-System in Wake	Conditions or Identifiers	Rate Slow in Wake Time Frame	Max Effect small (limiting reactant)	Transport Limitations Potentially Significant	Not Important Question for Long Term Perturbations	Reason why not important for Long Term
$CO \rightarrow CO_2$	Exhaust and Wake	X				
$HC + O_3 \rightarrow$	Unburned HO			X	X	No catalytic chains
$NO + O_3 \rightarrow NO_2 + O_2$	Large NO_x engine emissions		X (O_3)		X	Initial ratio of NO to NO_2 not important in long run
$NO + O_3 \rightarrow NO_2 + O_2$	Very clean engine Max ambient O_3	X		X	X	Ditto, and O_3 in wake is small fraction of total O_3
$NO_2 + OH \rightarrow HNO_3$	Ambient OH		X (OH)			
$NO_2 + OH \rightarrow HNO_3$	Nonequilibrium OH prod. in max afterburn		? (OH)	X (in jet)		
$O_3 \xrightarrow[NO_x]{h\nu} O_2$	Photochem, catalyzed, ambient O	X			X	O_3 in wake small fraction of total O_3
$O_3 \xrightarrow[H_2O]{h\nu} O_2$	Photochem, H_2O catalyzed	X			X	H_2O catalysis unimportant
$NO \xrightarrow[O_2]{O} NO_2$	Exhaust and Wake	X			X	Initial ratio NO/NO_2 not important in long run
$SO_2 \xrightarrow[O_x]{h\nu} H_2SO_4$	Photochem. formation of particulates	X				
$SO_2 \xrightarrow[cat]{h\nu}$ smoke	Ditto, accelerated by NO_x + unburned HO unsaturates	X (excess NO)	X (-C=C-)			

Discussed below are several of the possible wake reactions and initial estimates of their progress.

$$CO + OH \xrightarrow{k_1} CO_2 + H$$

In hydrocarbon-air systems, this reaction is generally considered to be the only important CO oxidation reaction at high temperatures, and is responsible for CO conversion to CO_2 in the exhaust jet. Using the rate constant (Leeds evaluation) for the above reaction and a partial equilibrium assumption for OH, negligible CO oxidation should occur in the wake.

Computer integration of the full set of elementary chemical rate equations governing CO oxidation confirms the conclusion that CO oxidation is negligible behind the engine.

NO/O_3 and H_2O/O_3 Photochemistry

The maximum effect of O_3 reduction in the wake due to catalytic nitric oxide or water vapor cycles would be to remove all of the O_3 in the wake. This reduction would not affect the global O_3 balance. Moreover, wake residence times are generally not long enough for significant photochemical reactions to progress in this region. In the case of NO_x-catalyzed O_3 decomposition, for example, the half-time for O_3 destruction is at least one order of magnitude greater (slower) than the local wake residence time.

Photochemical Formation of Sulfate Aerosols

In the absence of hydrocarbons or NO, the photochemical conversion of SO_2 to H_2SO_4 is not appreciable in the wake regime, where about 0.01 percent of the SO_2 is converted to H_2SO_4.

In the presence of hydrocarbons and NO_x, the rate of aerosol formation is much greater. However, the aerosol formation does not begin until NO is oxidized to NO_2, and complete oxidation of NO is not expected in the wake.

$NO + O_3 \rightarrow NO_2 + O_2$

This reaction is of interest in the far wake where the combined effects of wake dilution and diffusional O_3 transport may lead to NO and O_3 concentrations of similar magnitude. In the stratosphere, half times for this reaction appear to be about 5 minutes. Thus, this reaction may have some significance in the far wake regime.

$NO_2 + OH \rightarrow HNO_3$

Ambient levels of OH in the wake are not large enough to form significant amounts of HNO_3 in the wake regime. With the engine at maximum afterburn, OH levels may be sufficient to cause the engine to emit some nitric acid.

Reactions of Unburned Hydrocarbons with Ozone

Conceivable reactions of hydrocarbons with NO_x and/or ozone do not appear to give rise to catalytic cycles. Therefore, the maximum ozone reduction by these reactions cannot exceed the total hydrocarbon effluent, and is therefore insignificant. The direct reactions with ozone can readily be shown to be noncatalytic. Reactions of hydrocarbons with NO_x which can affect ozone fall into two classes, one which interchanges odd nitrogen among the organic radicals, and one which results in a net conversion of NO to NO_2. If the latter class could accelerate the $NO \rightarrow NO_2$ conversion rate, subsequent photolytic steps are conceivable which might affect ozone levels. However, no cyclic (chain) processes through which HC increases the $NO \rightarrow NO_2$ rate appear; rather, the processes appear to degrade the HC species until they are consumed. At most there result as many $NO \rightarrow NO_2$ conversions as there are carbon atoms in the system, with the further limitation that methane cannot participate in this sequence.

$NO + O \rightarrow NO_2$ and $NO + NO + O_2 \rightarrow 2NO_2$

The question of NO oxidation in the engine exhaust nozzle and aircraft wake may be significant. For example, short-term nitric acid formation in the lower stratosphere may depend on the partition of NO_x between NO and NO_2 in the final wake. Initial estimates of the progress of these reactions in the wake give minimal conversion of NO to NO_2. However, it is not inconceivable that under some engine operating conditions, there is significant oxidation in the engine exhaust nozzle and near wake field.

THE EFFECTS OF CONCENTRATION INHOMOGENEITIES ON MEAN CHEMICAL RATES

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Dr. Glenn Hilst presented to the joint panels a discussion of higher order correlation terms as they may affect chemical rates. The following more primitive argument considers only first-order correlations between spatially fluctuating reactant concentrations and parameterizes these in terms of a specific (log-normal) probability distribution.

Specifically, suppose $dc/dt = k_n c^n$,

let $$\xi = \frac{1}{\sigma} \ln \left[\frac{c}{\bar{c}} \right]$$

and assume $$P(\xi)\, d\xi \cong e^{-\xi^2/2}\, d\xi / \sqrt{2\pi}$$

then $$\overline{\frac{dc}{dt}} = k_n \bar{c}^n \cdot e^{n^2\sigma^2/2}$$

If $n = 2$ and $\sigma = \begin{bmatrix} 0.1 \\ 1.0 \end{bmatrix}$, then the exponential factor is $\begin{bmatrix} 1.02 \\ 7.3 \end{bmatrix}$, respectively.

If this perhaps too simple formulation is at all helpful, the next question is, what is the σ which usefully describes the stratospheric concentration fluctuation for the rate limiting species? If σ is $O(0.1)$, then concentration fluctuations would appear not likely to affect the stratosphere's macrochemistry, and conversely, if σ is $O(1.0)$. My unsupported guess is that the former case applies.

Conceptually, however, σ is capable of experimental measurement. Moreover, the moments of real concentration fluctuations are in principle measurable, and the real distribution of the fluctuations could be integrated explicitly.

Aerosols, An Influence Upon Stratospheric Chemistry?

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The aerosol content in and above the stratosphere may plausibly affect the stratospheric chemistry in at least three ways: (a) the optical scattering and absorption of solar radiation by aerosols may diminish the radiation available for photochemical processes. (b) the aerosol's surface area may promote heterogeneous chemical reactions at rates exceeding those of homogeneous processes. (c) the aerosol may serve as a relatively inert sink of stratospheric constituents which might otherwise participate in the chemical stew.

Let us discuss each of these suggestions in turn.

(a) Recent measurements (Hodge, 1972) using both nephelometry and stellar extinctions by astronomical, photoelectric photometry have indicated that:

1. In remote locations the scale height of tropospheric aerosol extinctions (3-4 km) is about half the scale height of the molecular constituents;
2. At Mauna Loa (altitude about 3.5 km) nephelometric scattering coefficients vary from 1.01 to 1.10 times Rayleigh scattering at 5000 A on the clearest days. This corresponds to an aerosol burden of a few (1-10) x 10^{-6} gms/m^3.

3. Astronomical measurements taken at the same site and times are consistent with the nephelometric data. That is, no denser aerosol column was discovered between Mauna Loa and the top of the atmosphere. The magnitude of Rayleigh extinctions is approximately 10% at 5000 A for one air mass.

Recent balloon sampling of the stratospheric aerosol (Lazrus, 1971) indicates local aerosol concentrations as high as 0.1×10^{-6} gm/m^3.

Together, these observations suggest that aerosol extinctions may be neglected safely with respect to Rayleigh scattering, for purposes of estimating photoeffective solar fluxes, near 5000A. Because Rayleigh scattering varies with wavelength approximately as λ^{-4}, this conclusion is also probably valid in the ultraviolet, where the Rayleigh extinctions should amount to about $2^4 \times 10\% = 160\%$. However, at 30 km, where ozone's mixing fraction is greatest, the air mass is about 0.1. Additionally, half of the Rayleigh-scattered flux is directed forward and is thus still effective in promoting photochemistry. All together, these numbers suggest perhaps a 5-10% diminution of effective solar flux due to non-absorptive scattering by the atmosphere, a value which should perhaps be included in our models, but which is about as large as uncertainties in the solar flux measurements themselves.

Finally, for simplest (Chapman) photochemical models, local ozone concentrations vary as the active flux to the 1/2 power, and considerable self healing occurs for

the stratospheric ozone column as a whole upon introduction of small perturbations in the solar flux intensities. Consequently, it is likely that neither aerosol nor molecular-Rayleigh scattering effects are very significant to computations of the earth's ozone budget.

(b) May heterogeneous chemistry significantly affect the stratospheric kinetics? Quite possibly. By dimensional analysis, it can be argued that characteristic times for gas diffusion to an aerosol's surface, T, should be

$$O\,(L^2/(K\phi))$$

where: L is some characteristic length describing the aerosol's size distribution (for example, a mean radius); ϕ is the ratio of the volume of the stratospheric aerosol's condensed phase to the dispersed volume; and K is a diffusivity. Separately, these factors are about 3×10^{-7} meters, 10^{-13}, and 0.1-1 meters ^{2}/sec, respectively. Thus, T should be O (1-10 sec). Therefore, if the surface's catalytic efficiency for some unspecified reaction approaches unity, the characteristic times for heterogeneous chemistry may be comparable to, or shorter than, those for many important homogeneous reactions. Some experimental evidence now bears on this speculation, as a negative correlation has been reported (Rosen, 1966) between the stratospheric aerosol and local ozone concentrations. We should be cautious, however, about mechanistic inference from correlation analysis; many traps abound from hidden variables.

(c) The last question is whether the stratosphere's aerosol inventory acts significantly as a chemical prison to sequester otherwise active chemical constituents. A quick partial answer is: clearly not for those constituents whose ambient gas concentrations appreciably exceed the stratospheric aerosol's mass concentration of 10^{-7} gm/m^3 ($\cong 10^9$ molecules /cm^3 $\cong 10^{-8}$ stratospheric mixing fraction, for molecular weight = 60). Many interesting and chemically significant stratospheric species are present at concentrations less than this value. However, to be influential these must probably be reactive, and thus catalytic effects appear to be potentially more significant than storage.

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Aerosols, An Effect Upon the Atmospheric Radiation Balance?

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Potential effects upon climate due to stratospheric aerosol concentration changes produced by SST effluents must be considered. The trend toward warming or cooling can be postulated to depend on the ratio of optical absorption to backscatter as well as on the earth's albedo. Models are being developed to handle aerosol multiple-scattering and absorption effects, and aerosol optical properties must be reliably estimated. Laboratory optical experiments on controlled aerosol distributions as well as on selected air samples are desirable sources of information. Photometric data which have already been collected must also be analyzed. In the visible wavelength region, aerosol contributions to the total optical extinction can be determined.

Of interest is the photometric data collected by Bener at Davos in the UV wavelength region. Aerosol contributions to total extinction can be estimated from Bener's measurements of direct solar radiation. Aerosol scattering properties may then be estimated from his sky-radiation data using a multiple scattering code to solve the radiation-transfer equation.

NUMERICAL ANALYSIS

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The space-time evolution of chemical species is given by the system of equations

$$\frac{\partial C_i}{\partial t} = \nabla \cdot \vec{V} C_i + F_i\,(\vec{C}\,(\vec{x},t)) - C_i R_i\,(\vec{C}\,(\vec{x},t)) + S_i\,(\vec{x},t) + H_i\,(\vec{x},t) \tag{1}$$

where $C_i = C_i\,(\vec{x},t)$ is the mean concentration of species i at position $\vec{x}$ and time t, $\vec{V} = \vec{V}\,(\vec{x},t)$ is the velocity, F_i is the local production rate by chemical reactions evaluated for the mean concentrations, $C_i R_i$ represents the corresponding destruction terms, $S_i\,(\vec{x},t)$ is the local source term (e.g., input due to aircraft effluents), and $H_i\,(\vec{x},t)$ is a term representing departures from mean variables. $\vec{C}$ is the vector whose components are C_i.

The term $\nabla \cdot \vec{V} C_i$ is specifically dynamic. This term has been and is the concern of fluid dynamicists. In the absence of transport, system (1) reduces to

$$\frac{\partial C_i}{\partial t} = F_i\,(\vec{C}\,(\vec{x},t)) - C_i R_i\,(\vec{C}\,(\vec{x},t)) + S_i\,(\vec{x},t) + H_i\,(\vec{x},t) \tag{2}$$

Chemical kineticists have traditionally omitted the term $H_i(\vec{x},t)$. The preceding section entitled "The Effects of Concentration Inhomogeneities on Mean Chemical Rates" considers it more specifically. If the term $H_i(\vec{x},t)$ is omitted, there remains a system that is decoupled in space-time. For each space point a set of independent ordinary differential equations results.

The problem of modeling the stratosphere involves, in principle, solving the full simultaneous set of equations, (1). In practice, this degree of generality is presently beyond the hope of solution. For this reason, models are currently split into strictly dynamic and strictly kinetic parts. Associated with the dynamics is a characteristic time, t_d; associated with the kinetics is a characteristic time, t_c. Disparities in t_c vs t_d suggest simplifying approximations, e.g., alternately solving the dynamics for frozen chemistry and then the chemistry for frozen dynamics, and iterating the alternation. Depending on the details of the problem, either the dynamics or the chemistry may dominate and therefore require the greater effort. A particular difficulty associated with the chemical kinetics, namely stiffness, has been only somewhat resolved at the practical level in the last five years. This difficulty requires further effort. It is presently known that appropriate numerical integration methods for this class of systems must be implicit for stability and highly accurate locally to prevent error accumulation.

There are several numerical integration methods now available which are suitable for stiff systems. For a given problem, the choice among these must be based upon computing efficiency, i.e., speed, and memory storage demand. Perhaps the most powerful method for general use is Gear's method. The major limitation of this method, and most other implicit methods, is the frequent need for costly matrix operations. This places a premium on restricting the number of species.

Because of the crucial role that the chemical subproblem plays in the overall modeling problem, the choice of integration method merits careful

consideration before a final choice is made. A poor choice can adversely affect the entire computational problem since the chemical problem constitutes a deeply nested inner loop in a multiply nested problem. Apparently, small time losses in this inner loop can lead to profound losses in the overall problem.

Methods and possible shortcuts, e.g., the steady state approximation, also require testing to validate their accuracy. Accuracy and reliability can be examined, for a given numerical integration method, by testing the method on systems which have known analytic, but highly nonlinear solutions. A number of these are known, and a worthwhile preliminary step is to take advantage of them.

Finally, the development of new nonlinear algorithms, of which an infinity of possibilities exists, merits consideration.

STRATOSPHERIC MEASUREMENTS

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In collaboration with the reaction rate sub-panel, the following constituents are found to be constituents important to the stratospheric chemical state:

O, $O(^1D)$, O_3

H, H_2, OH, H_2O, HO_2, H_2O_2

N, NO, N_2O, NO_2, NO_3, N_2O_3

CO, CH_3, CH_4, CHO, CH_2O

SO_2, HNO_2, HNO_3 .

In addition, measurements of solar flux, particularly in the ultraviolet, and of aerosols are important. Not all of these constituents and parameters can be measured currently.

Of the above, the following are given highest priority for measurement:

O_3, OH, H_2O, NO, N_2O, NO_2, HNO_3, CO, CH_4,
aerosols, and ultraviolet flux from 2000A to 3400A.

There exists an urgent need to measure the concentration of NO below 40 km, because of the important role this gas plays in the destruction of ozone and because of the complete lack of such data. Photoluminescent techniques

used to observe the NO concentration in the upper atmosphere can be adapted to probe the stratosphere. Adequate chemiluminous techniques are also being developed.

As a first step natural background profiles of the more important unknown species should be obtained. It is most important that as many species as possible be measured simultaneously, and at the same location, along with solar flux and basic meteorological observations. The next step should be measurements at different latitudes and seasons.

The following techniques are believed to be suitable presently for the respective constituents:

O_3	spectrometry, electrochemistry, chemiluminescence
H_2O	dew point determination, IR radiometry
NO	photoluminescence, chemiluminescence
NO_2	IR radiometry
HNO_3	IR radiometry, filter absorption followed by colorimetry
CH_4, N_2O	direct sampling and gas chromatography
CO	HgO reduction/Hg spectroscopy, IR spectroscopy
Aerosols	direct sampling with filters and impactors, in situ photometric devices, lidar, twilight measurements.

Photoluminescent techniques for measuring CO are within the state-of-art. Recent OH measurements in the upper atmosphere by photoluminescent methods hold promise that such techniques may be extended to measure concentrations in the stratosphere. SO_2 also may be measurable by similar photoluminescent techniques. Infrared techniques currently being developed should prove to be of value in measuring H_2O, CH_4, CO, and NO.

Mass spectrometric techniques, if adaptable to the stratosphere by 1974, may afford possibilities for measuring many species simultaneously.

INTERACTIONS AMONG CHEMICAL MODELERS, FLUID-DYNAMIC MODELERS, AND MEASUREMENT MAKERS

Interactions Between Chemical and Fluid Models

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Input from both photochemical and fluid dynamic modelers will be necessary in order that ozone distributions, and the resulting amount of UV radiation reaching the ground, can be accurately calculated.

At present it is difficult to assess the relative contributions of photochemistry and atmospheric transport to observed ozone concentrations as a function of height, latitude and season. Depending upon which mechanism is important, the projected SST fleet may have very different effects. If the Chapman theory is valid, most of the ozone is produced in equatorial regions in the summer months and transported poleward. If this is the case, SST effluents—deposited predominantly at mid-latitudes in summer months—may have minimal effects on ozone concentrations due to the large transport of ozone poleward from equatorial regions. This large transport through mid-latitudes might tend to mask any smaller changes in ozone concentrations occurring there as a result of SST flights.

On the other hand, NO_x catalytic reactions with odd oxygen atoms may be dominant. If enough N_2O is transported (or diffused) from the troposphere into the stratosphere, the reaction

$$O(^1D) + N_2O \rightarrow 2NO \qquad (1)$$

must be considered. SST effluents may now have a totally different effect. Since (1) is photochemically controlled through

$$O_3 + h\nu \rightarrow O(^1D) + O_2 \tag{2}$$

there is the possibility that there will be less O_3 where there is more solar radiation (in contrast to the Chapman model). In this case, most O_3 may be created at mid-latitudes and any change due to SST effluents could be accentuated.

The resolution of this combined photochemical transport problem can be sought on several levels. Efforts will be required to include important photochemistry in stratospheric transport models. More rapid answers also can be sought by solving competing photochemical schemes first with no transport. Then the transport necessary to fit observed ozone concentrations can be calculated. If the resulting transport can be tagged as realistic, the photochemical reactions corresponding to the given transport scheme could be considered dominant to a first approximation.

INTERACTIONS AMONG CHEMICAL MODELERS, FLUID-DYNAMIC MODELERS, AND MEASUREMENT MAKERS

Model Interactions with Measurement Programs

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Models, both chemical and fluid-dynamic, should be used to assist measurements. Models should help specify conditions where measurements will be most sensitive to desired parameters and least sensitive to other, extraneous parameters. Where this is not possible, models can be used to correct for extraneous parameters as they influence measurements.

Models can assist interpolation in space and time between sparse measurements, and they can assist averaging sparse measurements (over the globe, or zones, or time, etc.).

Models can specify just which parameters most sensitively affect computed O_3 and thus establish a hierarchy of priorities for measurement programs.

IV. SUMMARY AND CONCLUSIONS

How close did the Asilomar workshop come to meeting its objectives? As discussed in the Introduction, these objectives were to:

- Review ongoing and planned activities pertinent to the CIAP objectives
- Foster interaction between the chemical and fluid-dynamic modelers
- Determine which, if any, of the current models of the atmosphere are best suited to CIAP needs and, if necessary, define for development new models directed specifically at the CIAP problem.

The first two objectives clearly were met by the workshop.

Participants in the workshop gained an understanding of the basic problems resulting from the wide range of time and spatial scales that must be considered by CIAP modelers. Available models with potential application to CIAP were reviewed.

Interaction between the two computational modeling groups was started. Means to facilitate this interaction, one example being a periodic newsletter, are being developed.

It generally was concluded that our present models require substantial refinements and extensions to be applicable to the CIAP problem. A hierarchy of models (such as those listed in Table 4.1) may be used to address different aspects of the problem; none of them may be expected to accomplish complete simulation by 1974. CIAP must depend, therefore, upon estimates based upon incomplete simulation.

The third workshop objective was only partially met. Since it is unlikely that a three-dimensional, time-dependent model with fully interactive chemistry and fluid-dynamics can be developed to fulfill CIAP needs by the 1974 deadline, appropriate simplified models directed specifically at the CIAP problem must be defined for development—and quickly.

Furthermore, increased efforts are essential to coordinate the activities of modelers and experimenters addressing the CIAP problem. This is especially true since simplified atmospheric models may rely heavily upon parameterized observational and experimental data.

In order to (a) define for development atmospheric models directed specifically at the CIAP problem, and (b) explore interactions between CIAP modelers and experimenters, a second CIAP Computational Modeling Workshop will be held in September 1972.

TABLE 4.1—CLASSES OF COMPUTATIONAL MODELS

Model Dimensionality	Typical Examples	Usefulness
Chemical reactions only, no dynamics	Univ. Washington (Harrison) V.P.I. (Levine)	Initial survey of sensitivity of chemical reactions
One-dimensional with parameterized transport	NOAA (Shimazaki) NASA Ames (Whitten) LMSC (Hoshizaki)	Surveys of sensitivity of chemical reactions using parameterized diffusive transport Studies of wake dynamics
Two-dimensional with parameterized dynamics	LLL (MacCracken) NOAA (Shimazaki) LMSC (Hoshizaki)	Effects of diffusive transport on chemistry Studies of wake dynamics
Three-dimensional with dynamics	GFDL (Mahlman) MIT (Cunnold) NCAR-Colorado (Washington, London) UCLA (Mintz)	Studies of dynamics affecting chemistry, but not dynamics affected by chemistry
Two-dimensional with interactive dynamics and chemistry	LLL (MacCracken)	Study of dynamics and chemical interaction of limited scope
Three-dimensional with interactive dynamics and chemistry	(Post-1974 only)	Complete atmospheric simulation

APPENDIX A

PLANNING A PROGRAM FOR ASSESSING THE POSSIBILITY THAT SST AIRCRAFT MIGHT MODIFY CLIMATE

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planning a program for assessing the possibility that SST aircraft[1] might modify climate

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Some background

It has been man's custom to pursue technological opportunities whenever they have promised short-term benefits. All too seldom have long-term disbenefits been predicted or considered in decision-making. Nonmonetary disbenefits—particularly effects on ecology and on the structure of society—have been neglected in our headlong pursuit of technology's promise.

This has got to stop! The time has come, and passed, when we *dare* venture forth with our new technology until we have assessed, as carefully as we know how, its potential effects on *all* aspects of our quality of life—and that of our great-great-grandchildren.

Societal effects may be nigh impossible to predict, for our understanding of human behavior is badly wanting. But we must try; and when we fail we must try again, until we learn how.

In contrast, we should do much better at predicting the *physical* effects of what we do. Great effort may sometimes be required, but the basic physical laws are dependable altogether, and we pride ourselves in our understanding of them. From here on, meticulous technological impact assessment of every venture we propose is a must. Anything less is inexcusable.

Transportation and the way men live

In the spectrum of human activities, transportation is a prominent example of the dynamic interaction between technological pursuit and the quality of men's lives.

Before there was transportation men lived in a very limited world. Each did nearly everything for himself, and existence was typically meager and dearly won (a situation often with much to recommend it). Gradually, transportation—boats and wagons—brought men a broadening of scope and began to weave an ever-increasing web of interdependence among them. With it came increasing specialization, a better material existence, greater opportunities for personal growth.

In recent history, transportation has had a predominant effect on the development of our social structure. Communities have grown in predictable fashion first around seaports, then along railroads. Then urban neighborhoods dutifully followed the deployment of streetcar lines.

Highways and airports introduced a new dimension, a new freedom. Now, which is cause and which is effect is less clear. But the interdependence is stronger than ever between transportation and community structure: stronger and less disciplined, and the result leaves much to be desired.

Today transportation is the very lifeblood of our economy. Our transportation arteries permit the interrelationships that sustain us in these years of popular growth. But they have "just growed up," and the state of our social structures and environmental preservation show it! The burden for ameliorating this process, starting now, is upon us all, and particularly upon us in DOT.

The Department of Transportation was created by the Congress for the purpose of addressing the nation's transportation as a single system, for thinking through how that system can grow to a better balance to serve all the people, while providing comprehensive protection to the environment in which it serves; and for doing those things the federal government ought to do to help achieve those goals.

In the vanguard of tomorrow's transportation system is today's research and development; and in today's context this includes not only developing the technology of new systems—new vehicle systems, new traffic control systems, new terminal access, etc.—but the careful, highly technical *assessment of the effects* that technology may have, with provision for fully controlling those effects—noise, emissions, visual pollution, violation of our precious wilderness, and all others.

We must do this for every component we think of building into our total transportation system—urban people-moving and transit systems, high speed railroads, highway and dual-mode systems, and the whole spectrum of air transportation systems.

Transportation technology and new system development is my responsibility, and I promise you that environmental protection has a first priority with me.

A commitment to protection

We take very seriously the responsibility for assessing preveniently the effects that proposed transportation

[1] Speech at the AMS 51st Annual Meeting, San Francisco, Calif., 12 January 1971.

systems may have—for preventing ill effects when this can be done, and for preventing deployment of the systems themselves whenever it cannot.

During the past two years we have vigorously pursued this policy in the areas of pollution, safety, and land impact.

We are ruling that smoke emissions from aircraft must be eliminated and they will! We have established stringent noise limits for new aircraft—and they are *being* met! Automotive emissions are being reduced on schedule, and at the same time alternative power plants are being energetically pursued. We are developing technology and seeking legislation which will give oil spill law enforcement the teeth to do its job.

Highway safety has turned a corner: in 1970, despite another large increase in passenger miles, the number of highway deaths went *down*. Three federal initiatives—seat belts, safety-oriented highway configuration, and anti-alcohol campaigns, are largely responsible. And there is much more coming, like air bags to save people too stupid to buckle their seat belts, increasing effective anti-alcohol law enforcement techniques, and the experimental safety vehicle program to develop cars that are containers of fragile-cargo first, and pleasure buggies second. One goal is total survival in any 50 mph head-on crash—and it will be met!

An airport was stopped in the Everglades and a highway was routed around Franconia Notch by a very human Secretary of Transportation who understands the preciousness of the wilderness—and who will take a stand and not budge.

These are small steps in a very large task, and there is much yet to be done. But we are determined to press forward relentlessly in every area where transportation impacts, or bids to impact the environment that gives us life and spirit.

A thrilling event of our time is the widespread public awakening to the urgency of the problems of technology impact such as those I have described, and the broad support one can now count upon in addressing these problems. It has been of tremendous value to us in mounting a program of technology assessment and impact abatement in the whole area of transportation.

A case in point: the SST

A salient case in point—and the subject of this paper—is the possible use of the supersonic transport as one component of our transportation system.

In many ways the potential adverse effects of the SST are similar to those of other transportation systems. Whether it is produced here or abroad only, it will fly low in the vicinity of the airports it uses—although it will be less noticed than other aircraft because of its much superior climbout capability. Its engines will be noisy on the runway, and we will have to require—and we *will* require—that major action be taken to guarantee that they will be no noisier than other aircraft. Engine emissions will be similarly controlled to the tighter future standards of all aircraft.

The sonic boom represented an intrusion on our lives which we would not abide, and we could not abate. So we are outlawing it over our country—first by DOT rule, and soon I expect by act of Congress. (It has been suggested that this rule will not stand up, but I am certain that it will. The speeds of all commercial aircraft are already rigidly controlled to a few knots' tolerance—and this control will get more stringent each year, as our air traffic control system evolves.)

Economically and sociologically the SST is one more step in the sequence whereby men have become more interdependent—but it is a big step. I see the SST as no mere new toy for the jet set (although idle hangers on are everywhere). To the contrary indeed! Just as the subsonic jet—with its five-hour access between city pairs on opposite coasts—has welded this country into a single, highly-interdependent economic, political, and social structure, so would the supersonic aircraft for city pairs the globe around; and not old international cities only, but even more important, *new* ones. Most profoundly, in my opinion, it would bring the rich, poverty-stricken underdeveloped countries of the third world into the front row in the family of nations.

For, I believe, five-hour access is the key to frequent discourse between individual leaders at all levels, in government, in commerce, in universities. That is what truly, finally is making this one nation. That is what could make this truly, finally, one world!

But we must be very sure we know the price.

The SST and the climate

The above effects—physical and economic—of the SST are of like kind—if perhaps greater degree—with those of other transportation systems through history.

But in one respect the SST just might present a unique problem. It has been suggested that the high-volume operation of a large SST fleet might produce a sensible change in the climate.

At this point we do not yet know enough to predict whether this will happen. We have to *learn* enough, before we make any production decision on American SST's.

We are now planning, with deliberate urgency, a program to obtain the necessary additional knowledge. I want to share our thoughts about it with you today, because the nation—the family of nations—needs very much your help in designing this program and in interpreting its results. The stakes are high. We have got to do the job right.

I propose first to describe for you the characteristics of projected SST fleet operations. Then I will review some pertinent aspects of the nature of the atmosphere, the ways that have been suggested that it might be affected by SST operations, and what is still to be learned before we will know. Finally I will discuss our preliminary planning of a program to gain the neces-

sary additional knowledge, and how we propose to carry the program out.

I want to acknowledge here the excellent work of Dr. Richard L. Strombotne, a senior physicist on my staff, whose responsibility is the area of vehicle emissions and atmospheric protection. Dr. Strombotne has led our studies on climatic assessment and the preliminary program planning I am about to discuss with you. Dick is a very competent and extremely conscientious man, as those of you who know him will attest.

The Climatic Impact Assessment Program will be managed directly from my office under Dr. Strombotne's scientific guidance.

The nature of proposed SST operations

It may be useful for me to begin by describing the characteristics of present flights and of proposed SST operations.

At the present time, U. S. military and commercial aircraft operate above the tropopause daily. In the next few years, the European and Russian supersonic transports are expected to go into production and subsequently to operate in the stratosphere at about Mach 2. If this country were to proceed with a supersonic transport, its commercial operations might start late in this decade.

It has been assumed that, by the year 1985, approximately 500 supersonic transport aircraft would be in operation around the world, with heaviest traffic then over the North Atlantic. By then, transoceanic passenger miles alone are expected to equal today's *total* air passenger miles the world over. Of the postulated 500-vehicle fleet, perhaps 2/3 might be U. S. made and 1/3 foreign made. The U. S. aircraft would be powered by four GE-4 engines; the foreign aircraft by the equivalent of two GE-4 engines. (The fleet could be expected to expand with time.)

A typical flight profile for the U. S. SST has a cruise regime from about 60,000 ft at the beginning to about 70,000 ft at the end, well into the stratosphere. The aircraft are expected to be in the air an average of seven hours per day.

A GE-4 engine produces 65,000 pounds of thrust at sea level, 20,500 pounds at 60,000 ft. During a typical flight, the fuel consumption at altitude is expected to be about 22,600 pounds per hour per engine. For each engine, preliminary estimates indicate that the products of combustion during an hour of operation will be 29,000 pounds of water, 70,000 pounds of carbon dioxide, and very much smaller amounts of carbon monoxide, oxides of nitrogen, unburned hydrocarbons, and oxides of sulfur, and less than one pound of soot and other particles.

The engine emissions are distributed the length of the flight path. The wake vortices of the aircraft will expand to entrain the engine exhaust emissions in a few moments. For a time, the outer boundaries of the wake vortices define the volume in which the engine emissions are deployed. The wake vortices then break up and the exhaust emissions are subsequently dispersed and may be transported about in the stratosphere.

Suggested effects on the atmosphere

A number of mechanisms have been suggested whereby the exhaust gas constituents of aircraft flying in the stratosphere may alter the existing state of affairs.

Each exhaust gas constituent ultimately has a sink, a process by which it is eventually removed from the stratosphere. While in the stratosphere, however, each exhaust gas constituent becomes a new resident. It has also an opportunity to react with natural constituents of the stratosphere in the presence of the ambient solar and infrared radiation. There has been some concern about the possible resulting short- and long-term effects upon the radiation balance of the Earth, because changes that may occur in the stratosphere may in turn have some effect close to the surface. Specific mechanisms are described below.

Moreover, because its temperature profile increases with altitude, the stratosphere is relatively stable, compared with the constantly changing troposphere, and residence times are normally much longer in the stratosphere. Measurements of radioactive fallout from bomb tests and other sources indicate that a characteristic time for the transport of air through the stratosphere and back into the troposphere is in the range of one or two years.

Thus, there has been speculation that the SST emissions could lead to sensible changes in our weather and in the amount of biologically damaging ultraviolet light that reaches the surface.

The stratosphere itself, of course, contains the major atmospheric gases, oxygen and nitrogen, plus natural quantities of carbon dioxide, water vapor, ozone, aerosols, sulfates, nitrates and other trace gases and elements. In most cases, the amounts expected to be contributed by a future SST fleet are small in comparison with the amounts naturally present, but in certain instances they may not be small. Let us consider these constituents in turn, and the uncertainties concerning them that need to be resolved.

CO_2. In the stratosphere, CO_2 is present at a concentration of about 320 parts per million. Man's activities are estimated to add about 0.5% of CO_2 to the atmosphere annually. However, measurements carried out over the past 8 to 10 years indicate an annual increase of only about 0.2%. Thus the sinks are able to consume more than half of the annual increment.

Approximately 15.4×10^9 tons of CO_2 are produced annually by man's activities. The 500-plane SST fleet, operating seven hours per day would produce about 0.2×10^9 tons annually—an addition to the human contribution of about 2%.

There is perennial concern about the possible consequences of a continuous increase in atmospheric CO_2. To the extent that the "greenhouse" effect of CO_2 is

valid, the surface temperature should rise. However, the effect is not yet fully assessed because estimates of the effects of CO_2 to date normally have used a static, radiative model and have not taken into account the full range of atmospheric dynamical processes, as, for instance, evaporation and condensation of water vapor and cloud formation.

If it should be established that the increase of CO_2 in the atmosphere is a problem, the SST fleet, along with the rest of man's activities that produce CO_2, would be a part of that probem. I believe it is fair to say that CO_2 is not a peculiar problem of SST operations. The line of reasoning I have followed here is essentially that presented in the MIT Summer Study of Critical Environmental Problems. Their recommendation is that CO_2 be monitored globally, but not associated with concerns about effects of the SST.

Water vapor. Water vapor is ubiquitous in the atmosphere, it plays many roles. It has been described as the working fluid in the tropospheric heat engine, since it is a carrier of energy from the high-temperature energy source at the equator to the cooler temperature heat sinks at higher latitudes. In the form of clouds it reflects the Sun's radiative energy back upward, and the infrared from the Earth's surface back downward. It also acts as an absorber and emitter of infrared radiation in much the same way as carbon dioxide. In comparison with CO_2, water vapor in the stratosphere does not radiate as much energy, in part because it is present in much lower quantities.

As one data point, Mastenbrook of the Naval Research Laboratory has measured water vapor in the stratosphere by balloon-borne instrumentation from the Chesapeake Bay over a period of years. At about 20 km, his measurements indicate about 3 ppm of water vapor. Other investigators have measured from 2 to 20 ppm.

Water vapor is believed to be introduced into the stratosphere by high thunder clouds extending above the tropopause, by the upwelling of moisture-laden air at the equator in the Hadley cell circulation, and by the interchange of stratospheric and tropospheric air that occurs in mid-latitudes by the action of the jet stream.

The mechanisms for removal of water vapor are not quite so clear, although a widely accepted explanation is that the extremely cold air at the tropopause acts like a cold trap for the stratospheric water vapor.

Preliminary estimates of the new equilibrium water vapor concentration in the stratosphere, based upon 3 ppm as the present value, the 500-aircraft fleet described earlier, and a life-time of 1-1/2 years, lead to a global average of 3.2 ppm. Locally, near flight routes, the concentration would be higher, and would depend upon the number of flights and the mass transport rates in the region.

Either a local or global increase in stratospheric water vapor may have several different effects. First, there is the radiative effect of water vapor itself which could constitute a small "greenhouse" effect. It could lead to warmer surface temperatures and a cooler stratosphere. Next, there could be greater cloudiness in the stratosphere in those regions, such as Norway, where conditions are favorable for cloud formation. A third effect could be a reduction in ozone. The likelihood of these postulated effects is quite uncertain.

The estimates of the little greenhouse effect of added water vapor are subject to the same comments others have already made about the "greenhouse effect" of CO_2. The dynamics of the atmosphere must be included to understand the consequences.

Calculations of the distribution of ozone in a wet atmosphere show qualitative agreement with measurements in the real atmosphere, but not detailed agreement. There is sufficient uncertainty about ozone-water photochemistry to prevent accurate estimation at this time of the change in total ozone resulting from increased stratospheric water.

Moreover, our knowledge of stratospheric water vapor is inadequate to permit us to know whether the Naval Research Laboratory's measurements out of Chesapeake Bay are typical of the stratosphere as a whole.

It is clear that extensive measurement of the presence and movement of water vapor in the stratosphere, together with modeling and carefully correlated simulation, are of major importance.

Contrails. The low humidity of the stratosphere makes it unlikely that condensation trails, that is, contrails, will form and persist for more than a short time. Many observations (e.g., of military aircraft) bear this out. The regions of the stratosphere where contrails have the greatest chance of persisting are the cold regions above the equator and near Iceland during the winter months. Variations in the relative humidity upward from the mean would increase the probability of contrails.

Since stratospheric water vapor measurements are difficult and lacking in the regions of greatest probability, we are not in a position to predict completely the possibility of contrail formation.

Persistent contrails may affect radiation balance and therefore need investigation. It should be pointed out, first, that persistent contrails have not been observed in the stratosphere and, second, that most flights in the stratosphere do not now occur where such contrails are most likely.

Particulates. Aerosols or particulates are natural constituents of the stratosphere. Measurements by Junge and Cadle and others have shown the existence of aerosol layers in the stratosphere. Natural sources include meteorites and volcanoes. Violent volcanic eruptions have injected large amounts of particulates into the lower stratosphere. Some eruptions have caused a many-fold increase in particulate concentrations, notably, the 30-fold local increase caused by the eruption of Mt. Agung on Bali in 1963.

The presence of particulates affects the amount of direct and indirect sunlight reaching the surface and the infrared radiation characteristics of the stratosphere.

The net effect of particulates in the stratosphere is still unknown. Calculations of the radiative properties and radiative transport of particulates depend upon knowledge of optical properties of the particles and details of their size distribution. It is known that temperatures rose in some regions of the stratosphere after the Mt. Agung explosion by as much as 6 or 7C, but there was no apparent temperature change in the troposphere. It is possible that SST operations could increase temperatures in the stratosphere if enough particulates are produced in the form of soot and hydrocarbons, or from the gaseous products of combustion; but the effects in the troposphere may well be altogether negligible. We just don't know yet.

Thus, again, there are many uncertainties about the possible ultimate effects of particulates produced by SST fleet operations. These relate to the mechanisms by which particulates are formed naturally and are removed, the amount of scattering, absorption, and reradiation of particulate layers, and the consequences of temperature increases in the stratosphere. These are uncertainties that need to be resolved.

Nitrogen oxides. Nitrates have been detected on filter samples of stratospheric particulates, and HNO_3 has been detected optically. The observations are relatively recent. Consequently, there is much to learn about the natural production of nitrates in the stratosphere.

There has been speculation about the possibility of a smog layer in the stratosphere or a reduction in atmospheric ozone as the result of SST engines producing oxides of nitrogen. NO reacts with ozone to form NO_2 and O_2 in a well-known chemiluminescent reaction. NO_2, in turn, may react with water and other stratospheric constituents to form HNO_3 and particulates. Our understanding of the possible role and significance of oxides of nitrogen is primitive in comparison with what is known about water vapor, ozone, and carbon dioxide.

Carbon monoxide. Carbon monoxide has many natural sources, including, apparently, the oceans. The natural sinks for CO are not as well-known. Measurements by Junge indicate that the stratosphere is a sink. Others are also possible. The stratospheric reactions involving carbon monoxide are becoming better understood.

The climatic impact assessment program

The SST Climatic Impact Assessment Program will be conducted from my office, as noted above. Its objective is to make the measurements, develop the models, and perform the analyses that are necessary to complete a comprehensive assessment of the possible effects of projected commercial supersonic transport fleet operations. This is to be accomplished prior to a national decision regarding production of SST's by this country.

Much of the necessary information has already been developed by meteorologists, atmospheric scientists, engine manufacturers, and others in previous years. Moreover, much pertinent work continues. There are, however, important gaps in the information base, and in our understanding of the atmosphere, that must be filled in during the course of this program and its time span. The necessary information would probably be generated over a period of years without this specific research program, but we intend to accelerate the process where the results are important to this decision.

Many organizations, both public and private, are already deeply involved in meteorological and atmospheric science programs, and many of you are heavily involved. We intend to draw upon the expertise and established capabilities of these groups in conducting this research program. We have, in fact, already had very valuable discussions with some of you about the content and direction of the research program. We shall continue to seek your contributions.

Content of the assessment program

It is convenient to describe the program in two main parts. The first of these involves monitoring the atmosphere *per se*. The second encompasses the logical structure that follows the time history of the engine exhaust emissions through to their ultimate consequence for climate.

The atmospheric monitoring program element is to make the measurements on water vapor, particulates, and trace gases that are required to project the effects of SST operations. These measurements will extend our knowledge of their geographic and seasonal variability. We will emphasize stratospheric measurements. We anticipate the use of RB57F and other aircraft and of balloons for *in situ* measurements. We will use also ground-based remote sensors, such as radar and light radar or lidar. Existing instrumental technology will be used to the greatest extent practical in the interest of truly accelerating the measurement program. Thus, frost point and infrared hygrometers, particle filters and nephelometers will almost certainly be prominently used. Many of the necessary measurements are being made now by other organizations. We would hope to expand the network of measurement stations by working through these organizations.

The second main strand of the research program is focused upon the time history of the engine exhaust emissions and their effects. It consists of three main program elements, labeled as engine emissions, photochemistry, and geophysical modeling.

Engine emissions. We should recall that the SST engines are still being developed. From the engineering design and preliminary tests we can be pretty confident about the amounts of water vapor and carbon dioxide that will be produced under various operating conditions. The amounts of oxides of nitrogen, hydrocarbons, and particulates produced will depend more upon details of the engine design and specific operating conditions. Thus, we will have to carry out a combination of tests and modeling studies to characterize more completely the engine exhaust emissions themselves. Work now underway will contribute to this effort.

General Electric will, of course, be making measurements of the developmental engines for the SST.

Where work planned by other agencies fails to provide the information needed for the assessment of SST effects, this program element will provide for the required measurements and modeling specifically. We also plan to examine fuel composition and fuel additives.

Atmospheric modeling (geophysical modeling). The atmospheric modeling program element will apply existing models of atmospheric dynamics and extend their capabilities to project the effects of engine exhaust emissions upon the stratosphere and upon conditions near the surface. Three regimes are of interest. They are the wake of the aircraft, the flight track region, and the atmosphere up to the stratopause.

Within the aircraft wake, the exhaust emissions have the most rapid variation of temperature, concentration, and composition. The chemical reactions that occur in this region need to be understood as functions of the ambient conditions, radiation field, and flow fields. With knowledge of the composition of the disorganized wake, one can follow the exhaust products into the next regime, that of the flight track region. A mathematical simulation of the wake region will be developed and validated by measurements to obtain the composition, or at least the concentrations, of selected chemical species.

Atmospheric motions disperse the exhaust emissions and reaction products from the flight path and reduce their concentrations further. There have been various estimates of peak concentrations of exhaust emissions in the flight track region. The MIT Study of Critical Environmental Problems, for example, estimated that the water vapor increase in that region due to SST fleet operations would be ten times greater than the increase globally. The factor of ten is a compromise between higher and lower estimates. A mathematical simulation for the transport of exhaust emissions out to about 1,000 kilometers is needed to provide input for the large global models. The model for the flight track region and beyond should include the chemical reactions, turbulence, and radiation environment.

Assuming that SST exhaust emissions and reaction products are dispersed widely throughout the stratosphere at a low concentration, a full assessment of their effects will require the use of a numerical model that simulates the dynamics of the atmosphere, such as the NCAR model or that of the Geophysical Fluid Dynamics Laboratory at Princeton. As elaborate and sophisticated as these models are at the present time, they are not suited for this task. Ideally, one would want a model that simulates the atmosphere up to the stratopause, is capable of keeping track of the important chemical species, and includes interactions between the atmosphere and the ocean. None of the existing models is capable of doing so. We anticipate supporting the evolution of present models in those directions at an accelerated pace. Some exploration of simplified models is also in order.

In all of these modeling efforts, validation is of high importance. The sensitivity of the model to variations of the input data and to its own structure must be known or established.

Photochemistry. The photochemistry program element will provide for the laboratory studies and theoretical investigations of chemical reactions in the stratospheric environment. Results of the studies will flow into the atmospheric models. We expect to extend the analysis of ozone interactions with water and nitric oxide (NO), for example. We also need to support the study of particulate formation from gaseous reactants.

It appears also that a deep look at the accuracy of rate constants and their temperature dependence for some critical reactions is in order. The reaction between OH and ozone in the chain by which water vapor removes ozone is one such. As we get deeper into the research program we expect that other reactions will be identified as being of possible significance and they too will be examined

In addition to this major thread of studies I have just described, we intend to investigate the possible generation of contrails in the most likely areas of the stratosphere and the effects of contrails upon thermal radiation balance as necessary.

We also intend to be sufficiently flexible in our approach to allow consideration of other topics that are brought to our attention, as such appear to be justified.

The program team

I would like to take just a few more minutes to discuss with you the likely roles of government, industry, and the universities in this research program. Earlier I said that we would be looking for the best talent and capabilities in the country to carry out the assessment of possible effects of the projected SST fleet upon weather.

We will be looking to university researchers for laboratory studies, theoretical analysis, and for advice about specific aspects of the research programs, consistent with their historic roles of free and independent inquiry. In some instances, university researchers have developed a unique capability to make specific measurements which may be of particular use in this assessment program.

Private companies and not-for-profit research organizations have an ability to marshall resources and complete defined projects that is essential for the successful conduct of this research program. Moreover, many have personnel, facilities, and experience that can provide an organized effort in the program elements I have been describing.

The principal government role in this program is to provide the overall management, to furnish such equipment as high altitude observational aircraft, and to focus unique resources of the government. Several govern-

ment agencies, NOAA, the Air Force, the Navy, the National Science Foundation, and NASA have a history of conduct and support of meteorological research and atmospheric science. Coordination of this research program with the ongoing activities of the other agencies is a continuing process.

Finally, we look to you, as responsible members of the meteorolological community, for guidance and assistance in carrying out an objective assessment of the effects of any future SST fleet. In fact, some of you have already been asked to help. Your contributions and your counsel are indeed vital to a proper program and a proper assessment.

Thank you again for the opportunity to discuss this important meteorological program with you, and to solicit your help.

APPENDIX B

ASSESSMENT OF CLIMATIC CHANGES DUE TO FLIGHTS IN THE STRATOSPHERE

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ASSESSMENT OF CLIMATIC CHANGES DUE TO FLIGHTS IN THE STRATOSPHERE

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ASSESSMENT OF CLIMATIC CHANGES DUE TO FLIGHTS IN THE STRATOSPHERE

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Abstract

The Department of Transportation Climatic Impact Assessment Program (CIAP) proposes to assess, by report in 1974, the impact of environmental and meteorological changes due to a world high-altitude vehicle fleet as projected to 1990. The study includes, in sequential steps, consideration of the nature of the stratosphere, the nature of the propulsion effluents of a projected 1990 fleet, the perturbation of the stratosphere resulting from the introduction of the propulsion effluents, the changes in the troposphere which result from the stratospheric changes, and the biological impacts of the changed troposphere.

This paper describes briefly the Department of Transportation's Climatic Impact Assessment Program and indicates some of the questions the study should address.

The objective of the DOT Climatic Impact Assessment Program (having the acronymn CIAP) is to assess, in form of a report in 1974, the impact of climatic changes resulting from perturbation of the upper atmosphere by the propulsion effluents of a world high-altitude aircraft fleet as projected to 1990.

The steps in the study are six in number. To be considered first is the nature of the stratosphere including its present distributions of temperature, constituents, motions and circulation.

By second step, proceeding concurrently, the nature of the propulsion effluents to be introduced at a date about 1990 is considered. This step takes into account the amount of the engine emission products and projections of routes and frequency, to be described as credible upper and lower bounds for the loading of effluents into the stratosphere. Both steps one and two may be, to some extent, experimentally verified.

A third step is a projection of the perturbed stratosphere described in terms of changed height distribution of constituent densities. The perturbed stratosphere is a result of the combination of the propulsion effluents considered in step 2 and the unperturbed stratosphere described in step 1. For this perturbation of the stratosphere one must consider more than 30 constituents, as well as changes in energy transport down and up through the stratosphere.

A fourth step is to consider the perturbations of the troposphere which may result from the changes in the stratosphere. These changes may include changes in heating, temperature, latitudinal temperature gradients, winds and precipitation, as well as changes in ultraviolet flux at the earth's surface.

A fifth step, based on the findings of step 4, is the determination of the physiological, biological and botanical effects of the tropospheric changes on man, animals and plants. These are the impacts with which people are concerned. In addition to the technological description of the biological environmental effects, there is needed as well a quantification in terms of economic measures, undertaken in step 6. These economic measures provide a measure for possible comparison with advantages, for instance, of travel in the stratosphere. Description of such advantages of stratospheric travel is not a subject of this study, however.

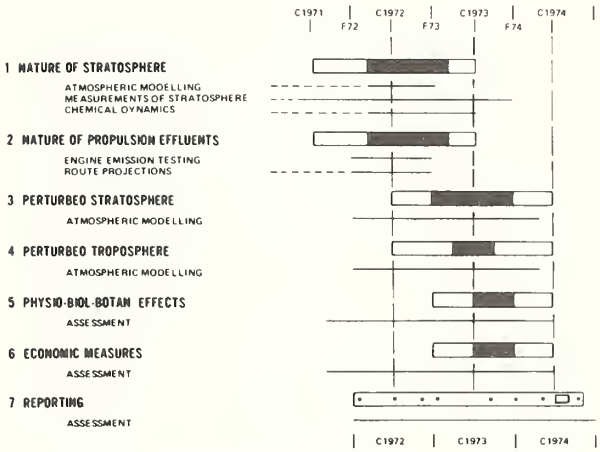


Fig. 1. CIAP program schedule.

In figure 1 is shown the schedule for the accomplishment of the major steps. The investigation of the geophysical nature of the stratosphere will continue throughout the program and will require the greater fraction of the funding which is available to DOT. This effort is a combination of measurements of the stratosphere, chemical dynamics and atmospheric modelling.

Step 2 is a concurrent study of the propulsion effluents and consists of both engine testing and route projections. For the accomplishment of the succeeding steps, the most useful results of steps 1 and 2 are those derived in calendar year 1972.

The accomplishment of step 3, the estimation of the perturbed stratosphere, involves atmospheric modelling, which is to be accomplished in the two years succeeding this summer, with the results most useful for succeeding steps being derived in calendar year 1973.

The studies of the perturbed troposphere, which is principally affected by the changes in solar radiation resulting from the perturbation of the stratosphere, also are accomplished with results expected to be derived chiefly in early calendar year 1973.

Estimation of the physiological, biological and botanical effects, which are the experienced impacts, is based on results of step 4. These estimations may be expected to be realized chiefly in the second half of calendar year 1973, to be expressed in terms both technological and to the extent possible economic.

As illustrated in Figure 1, the startup of the coordinated CIAP study occurred in FY-1972. The assimilation of the new and old data bearing on the problem of CIAP must be accomplished mainly in CY-1973, in order that the report may be drafted in CY-1974.

A seventh item, indicated also in the schedule, indicates the approximate times for major reporting milestones. These are shown by the asterisks to occur at intervals of about six months and to culminate in a final report at the end of CY-1974. Further details of this reporting schedule are discussed after a brief diversion.

The DOT Climatic Impact Assessment Program is dependent on work going on as the planned activity of more than 50 Government laboratories listed in Table 1. Such work,

Table 1. Agencies coordinating with CIAP.

AEC	**NOAA**	**N**
LASL	ARL	ONR
HASL	ERL	NRL
LLL	BL	
LBL	GFDL	**A**
BNL		WHITE SANDS
SANDIA		
ANL	**EPA**	
	DURHAM	**DOI**
NASA		MINES
AMES	**DOD**	
LEWIS	DNA	**HEW**
GODDARD SFC	DDR&E	NIH
GISS	ARPA	USPHS
HUNTSVILLE	AF	
	AEDC	**CEQ**
NSF	APL	
NCAR	CRL	**OST**
	AWL	
NBS	AWS	**NASC**
	AFTAC	

relevant to CIAP and particularly concerning steps 1 and 2, is being accomplished under the sponsorship of various agencies such as the Atomic Energy Commission, NASA, National Science Foundation, Department of Commerce, Department of Defense and others. Each of the laboratories listed on the Table is on its own initiative doing work recognized to be relevant to CIAP. While this work in general is necessary to the mission of and has been justified by the sponsoring agencies, none of the other agencies has as its particular goal the realization of a summary report in 1974 of the kind described here as a goal of the DOT CIAP study. The Department of Transportation intends therefore to exploit this relevant ongoing work to the maximum, and with the cooperation of the other sponsoring agencies, to introduce the results into the report of final CIAP conclusions. There is also ongoing work in Europe under the sponsorship of the developers of the CONCORDE and of the TUPOLEV 144 and also of other governments which is relevant and potentially a major contribution to the CIAP study. DOT hopes to introduce the results of such work in its report of findings. DOT also expects to sponsor new work and to exploit ongoing efforts in the universities and in industry. The Department of Transportation invites

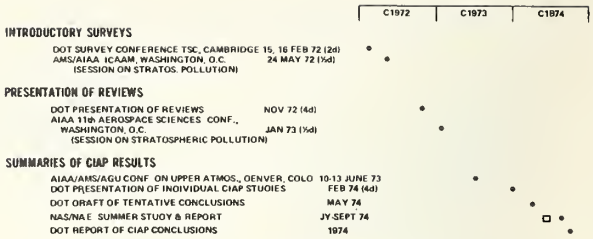


Fig. 2. CIAP reporting milestones.

proposals, within its budget limitations, for contributions relevant to the purposes of CIAP. DOT expects to take maximum advantage of ongoing programs supported by other agencies and governments and to use its limited funds to fill in the missing gaps required for a coherent report in 1974.

In Figure 2 are indicated the dates of some important reporting milestones. These reporting milestones represent activities, some under the direct sponsorship of the Department of Transportation, others under the sponsorship, we hope, of professional societies. As a follow-on to conferences of earlier years (SCEP and SMIC), a survey conference in February 1972 constituted an introduction. The purpose of the survey conference was primarily to introduce the breadth of the subject to principal participants, each of whom was invited by name to the conference in recognition of his special contribution in an area of scientific interest relevant to CIAP.

The AMS- and AIAA-sponsored International Conference for Aeronautical and Aerospace Meteorology (ICAAM) presented a one-day session concerned with stratospheric pollution on the 24th of May 1972, in Washington, D.C. This also served as an introduction in a more public way.

Although survey introductions are valuable, it is desirable that comprehensive, completely referenced review papers be prepared to cover topics for which adequate reviews do not currently exist. Examples of some that do exist are those critical reviews prepared by Elmar Reiter under AEC sponsorship. The Department of Transportation plans to organize the presentation of such comprehensive reviews within a time period adequate for their preparation, to be presented perhaps at the end of this year. A public forum for some of these reviews may be the AIAA Eleventh Aerospace Sciences Conference from the 10th through the 12th of January 1973, in which a half-day session is dedicated to atmospheric pollution.

The summaries of CIAP studies may be expected to appear in calendar year 1973. A conference suitable for public discussion of the studies, to be sponsored by the AIAA and possibly also by other technical societies, is planned for the summer of 1973. Also in February 1974 DOT will sponsor a conference for presentation of individual CIAP studies. These will provide the basis for the DOT first draft of conclusions to be prepared in May 1974. Inclusion of reviews of CIAP international meetings is being considered by the International Association of Geomagnetism and Aeronomy (IAGA) and the International Association of Meteorology and Atmospheric Physics (IAMAP).

The Department of Transportation is currently discussing with the National Academies of Science and

Engineering, the possibility of a 1974 summer study under the Academies' sponsorship, leading to a NAS/NAE report in the fall of 1974 of conclusions as to the impact of climatic changes. Such a study will have available as a primary data source all results of the CIAP effort. The final Department of Transportation report of CIAP conclusions, which will take the Academies' summer study report into consideration, is expected at the end of 1974.

The following figures illustrate conceptually the way in which various steps of the CIAP problem may relate to each other and may lead to conclusions concerning the impact of climatic changes. At this stage, these data only illustrate the nature of questions to be addressed by the study.

A proper first question is:

Q.1. "What are the general relations of radiation and dynamic control of the stratosphere, photochemistry, climatology and dynamics of the stratosphere?"

In Figure 3 are shown temperature-altitude profiles at several latitudes representative of January and mid-latitude Spring and Fall seasons. Also shown in Figure 3 are the

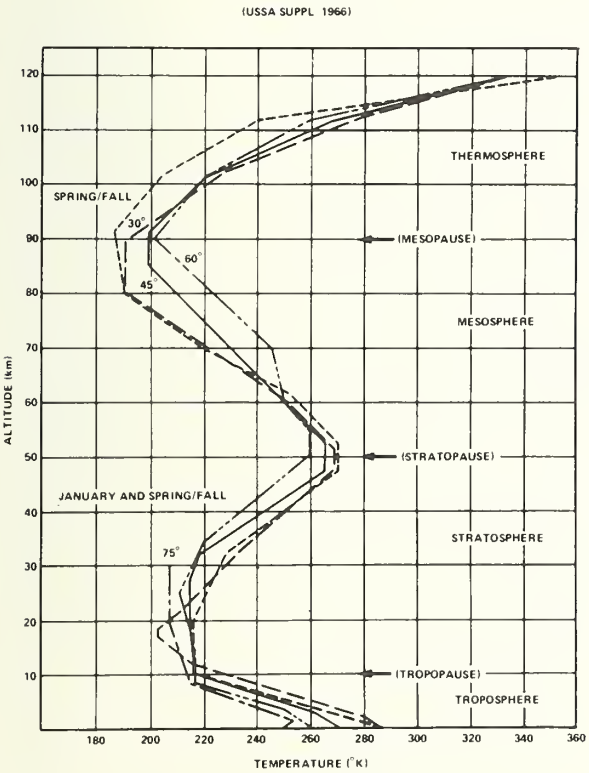


Fig. 3. Temperature-altitude profiles representative of January and mid-latitude Spring and Fall.

names of the atmospheric regions: the troposphere, below about 10 kilometers, the stratosphere, between the temperature minimum at the tropopause and the temperature maximum at the stratopause at about 50 kilometers, the mesosphere, which is the region intervening between the temperature maximum at the stratopause and the minimum above at the mesopause, and the thermosphere above the mesopause. The stratosphere, with which CIAP is concerned, is characterized by a positive temperature gradient, the principal effect of which is to inhibit vertical convective transfer of material within that zone. The temperature inversion is strongest above the altitude of 30 kilometers. However at latitudes greater than 30° the inversion is sufficiently strong above 10 kilometers to seriously affect vertical motions.

Other important questions relating to composition to be addressed by the CIAP program are:

Q.2. "What are the chemical constituents of the stratosphere?"

Q.3. "What are the important chemical reactions of the gaseous stratosphere?"

Q.4. "What are the relations between the gaseous chemistry and the aerosol chemistry?"

Some of the important constituents are shown in the plot of Figure 4 for the daylight atmosphere. These data are the work of Bortner and Kummler, 1969. The constituents shown represent only two thirds of the number of stratospheric constituents which we need to know for CIAP. Of particular interest in Figure 4 is the region between about 10 and 50 kilometers. As may be seen the constituents vary in mixing ratio from parts per million to parts per billion to one part in 10^{16}.

Important for consideration are the chemical reactions. Shown in the listing of Table 2 are gaseous chemistry reactions believed to be important and the uncertainties that have been associated with the rate constants (k) and photochemical absorption cross section (σ) associated with each listed reaction. This list, compiled by a workshop at Asilomar, California, meeting in April 1972, lists 63 reactions important in the stratosphere.

The constituents listed in Figure 4 may have great variability over the globe of the earth. In Figure 5 is shown the total ozone content in a column above the surface of the earth expressed in units of one thousandth of a centimeter, at standard temperature and pressure. On one day the total ozone varies over the earth from about 2 millimeters at STP to more than 6 millimeters. Since the radiative flux in the ultraviolet part of the spectrum which may drive important photochemical reactions is very sensitive to the vertical column of ozone, one may sense how important this variation may be.

Further questions to be addressed by the CIAP program include:

Q.5. "How well known are the chemical rate coefficients and which of these need further study?"

Q.6. "What are the aerosols naturally resident to be introduced and what are the relations between the atmospheric chemistry and the contained aerosols?"

Of the important reactions of the gaseous chemistry which are listed in Table 2, 27 are required by CIAP to be further evaluated as to chemical reaction rate coefficient (to an uncertainty less than factor 2) and 5 are required to be further evaluated as to photochemical absorption cross section in the ultraviolet and visual spectral regions.

Part of the CIAP study is concerned with pollutants which may be introduced by the 1990 world-wide fleet. These pollutants affect the stratosphere by complicated chemistry so one may ask the questions:

Table 2. Reactions and confidence limits for their rate constants.

Note: An updated version of this Table appears as Table 3.1 on pages 3-5, 3-6 of the main text of this workshop report.

Reaction No.	Reaction	Confidence Statement
	I. Basic Chapman Mechanism	
1	$O_2 + h\nu \rightarrow O + O$	$\sigma \pm 20\%$
2	$O + O_3 \rightarrow O_2 + O_2$	k well known
3	$O + O_2 + M \rightarrow O_3 + M$	k well known
4	$O_3 + h\nu \rightarrow O(^1D) + O_2$	$\sigma \pm 10\%$
	II. Destruction of Odd-Oxygen by NO_x	
5	$NO + O_3 \rightarrow NO_2 + O_2$	$k \pm 20\%$
6	$NO_2 + h\nu \rightarrow NO + O$	σ
7	$NO_2 + O \rightarrow NO + O_2$	k + 20, −50%
	III. Removal of NO_x	
8	$OH + NO_2 \xrightarrow{M} HNO$	k factor of 2, function of pressure
9	$OH + HNO_3 \rightarrow H_2O + NO_3$	k factor of 2
10	$HNO_3 + h\nu \rightarrow H + NO_3$	σ factor of 100
11	$\rightarrow OH + NO_2$	
	IV. NO Production	
12	$N_2O + O(^1D) \rightarrow NO + NO$	k factor of 2
13	$\rightarrow N_2 + O_2$	k factor of 2
14	$N_2O + h\nu \xrightarrow{\sigma_I} N_2 + O(^1D)$	$\sigma_I \pm 20\%$
	IV.A. Hypothetical Reaction Affecting NO	
15	$N_2O + h\nu \xrightarrow{\sigma_{II}} N + NO$	Important if $\sigma_{II} = .01$ $\sigma_I(N_2O + h\nu \rightarrow N_2 + O)$
16	$N_2(A) + O \rightarrow NO + N$	
17	$O(^1D) + M \rightarrow O(^3P)' + M$	$O(^3P)'$ indicates hot $O(^3P)$
18	$M + O(^3P)' + N_2 \rightarrow N_2O^* + M$	N_2O^* indicates excited N_2O
19	$N_2O^* \rightarrow NO + N$	
20	$\rightarrow N_2 + O$	
21	$\rightarrow N_2O$	
22	$O(^1D) + N_2 \rightarrow N_2O^*$	Slow
	V. Higher Oxides of Nitrogen	
23	$NO_2 + O_3 \rightarrow NO_3 + O_2$	k factor of 10^4 uncertainty at stratospheric T depending on activation energy = 7 ± 2 kcal.
24	$NO_3 + h\nu \rightarrow NO + O_2$	σ
25	$NO_3 + h\nu \rightarrow NO_2 + O$	σ small at peak, possibly important at lower wavelength
26	$NO_2 + NO_3 \rightarrow N_2O_5^*$	$k \pm 20\%$

σ – Photochemical absorption cross section
k – Reaction rate cross section

Table 2. (Cont.)

	V. Higher Oxides of Nitrogen (Cont.)	
27	$N_2O_5^* \rightarrow NO_2 + NO_3$	
28	$\rightarrow N_2O_5$	
29	$N_2O_5 + h\nu \rightarrow$	Products unknown, being worked on
30	$N_2O_5 + O \rightarrow$	
31	$N_2O_5 + OH \rightarrow$	
32	$N_2O_5 + HO_2 \rightarrow$	
33	$N_2O_5 + H_2O \rightarrow$	Slow in gas phase
	VI. $O(^1D)$ and O_3	
34	$O(^1D) + O_3 \rightarrow O_2 + O_2$	
35	$O(^1D) + M \rightarrow O(^3P) + M$	
	VII. Reactions Affecting OH, HO_2	
36	$OH + O \rightarrow H + O_2$	k factor of 2
37	$H + O_2 + M \rightarrow HO_2 + M$	k factor of 2; probably only important H reaction
38	$HO_2 + O \rightarrow OH + O_2$	k not known
39	$OH + O_3 \rightarrow HO_2 + O_2$	k factor of 10^3
40	$OH^\dagger + O_3 \rightarrow H + O_2 + O_2$	k depends on vibrational T; † indicates vibrational excitation
41	$OH + CO \rightarrow CO_2 + H$	k factor of 2
42	$O(^1D) + H_2O \rightarrow OH + OH$	k factor of 2
43	$O(^1D) + OH_4 \rightarrow OH_3 + OH$	k factor of 2
44	$O(^1D) + H_2 \rightarrow OH + H$	k factor of 2
45	$OH + HO_2 \rightarrow H_2O + O_2$	k factor of 2×10^{-10}, seems high
46	$OH + OH \rightarrow H_2O + O$	k factor of 2
47	$H + O_3 \rightarrow OH^\dagger + O_2$	k factor of 2
48	$HO_2 + HO_2 \rightarrow H_2O_2 + O_2$	k factor of 2
49	$H_2O_2 + h\nu \rightarrow OH + OH$	σ factor of 2
50	$H_2O_2 + O \rightarrow HO_2 + OH$	k factor of 2
51	$H_2O_2 + OH \rightarrow H_2O + HO_2$	k factor of 2
52	$HO_2 + NO \rightarrow OH + NO_2$	k probably high
53	$HO_2 + CO \rightarrow OH + CO_2$	k low, factor of 10
54	$OH + CH_4 \rightarrow H_2O + CH_3$	k factor of 2
55	$OH + H_2 \rightarrow H_2O + H$	k factor of 2
56	$OH + h\nu \rightarrow O + H$	σ very uncertain
57	$HO_2 + h\nu \rightarrow H + O_2$	σ very uncertain
58	$CH_3 + O_2 + M \rightarrow CH_3O_2 + M$	
59	$CH_3O_2 + NO \rightarrow CH_3O + NO_2$	
	VII. Reactions Affecting OH, HO_2	
60	$CH_3O_2 + CH_3O_2 \rightarrow CH_3O + CH_3O + O_2$	k factor of 2
61	$CH_3O + O_2 \rightarrow CH_2O + HO_2$	
62	$CH_2O + OH \rightarrow HCO + H_2O$	k factor of 2
63	$HCO + O_2 \rightarrow CO + HO_2$	k high; uncertain but sufficient

Q.7. "What are the emission species and mass rates at optimum power levels of the engines which will be flying at the stratospheric altitudes?"

Q.8. "What are the routes and frequencies of travel which are projected for 1990?"

These questions may be answered in terms of the output of and traffic by engines of present design (by tests of J-85 and the CONCORDE Olympus 598 engines) and of projected outputs of and traffic by engines conceivably to be developed in the period up to 1990 (as for hypersonic vehicles, for instance).

In considering the general dynamics of the natural stratosphere one may ask the questions:

Q.9. "How is the height distribution of radiation within the stratosphere determined?"

Q.10. "How is the radiation affected by the chemical and aerosol constituents of the atmosphere?"

In Figure 6 are shown values of absorption cross sections for ozone absorption in wave bands which are of interest to the CIAP study. It is necessary to know absorption cross sections in the ultraviolet for about fifteen photochemically sensitive constituents.

Other questions to be addressed by CIAP are:

Q.11. "What is the radiation within the stratosphere received from the sun, expressed in terms of mean and standard deviations?"

Q.12. "What is the radiation within the stratosphere received from below?"

These radiations are functions of the absorption cross sections as well as the densities of constituents above and below the altitude of particular interest and of the solar flux. In the case of the solar flux, the flux outside the

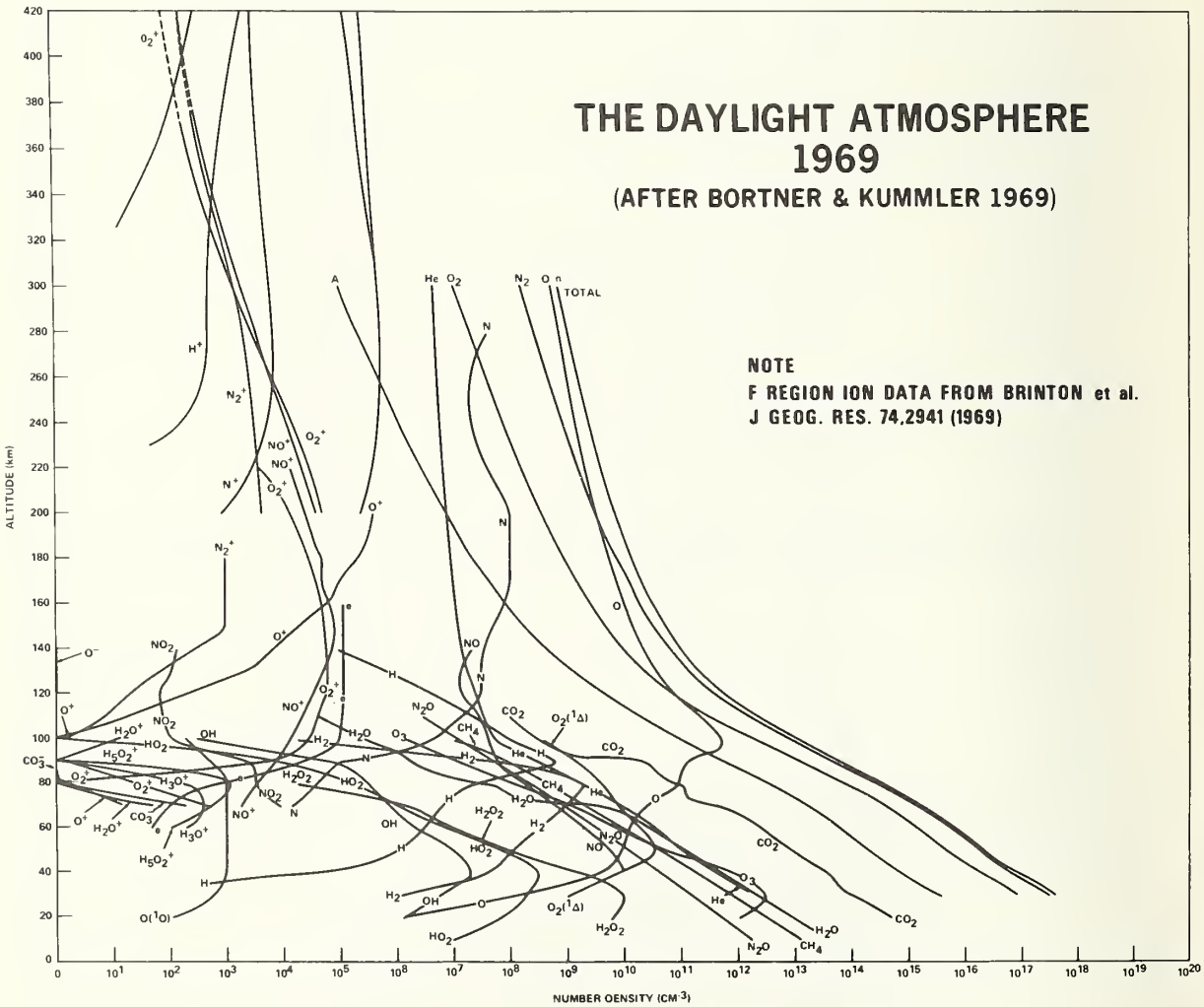


Fig. 4. Some important constituents of the daylight atmosphere.

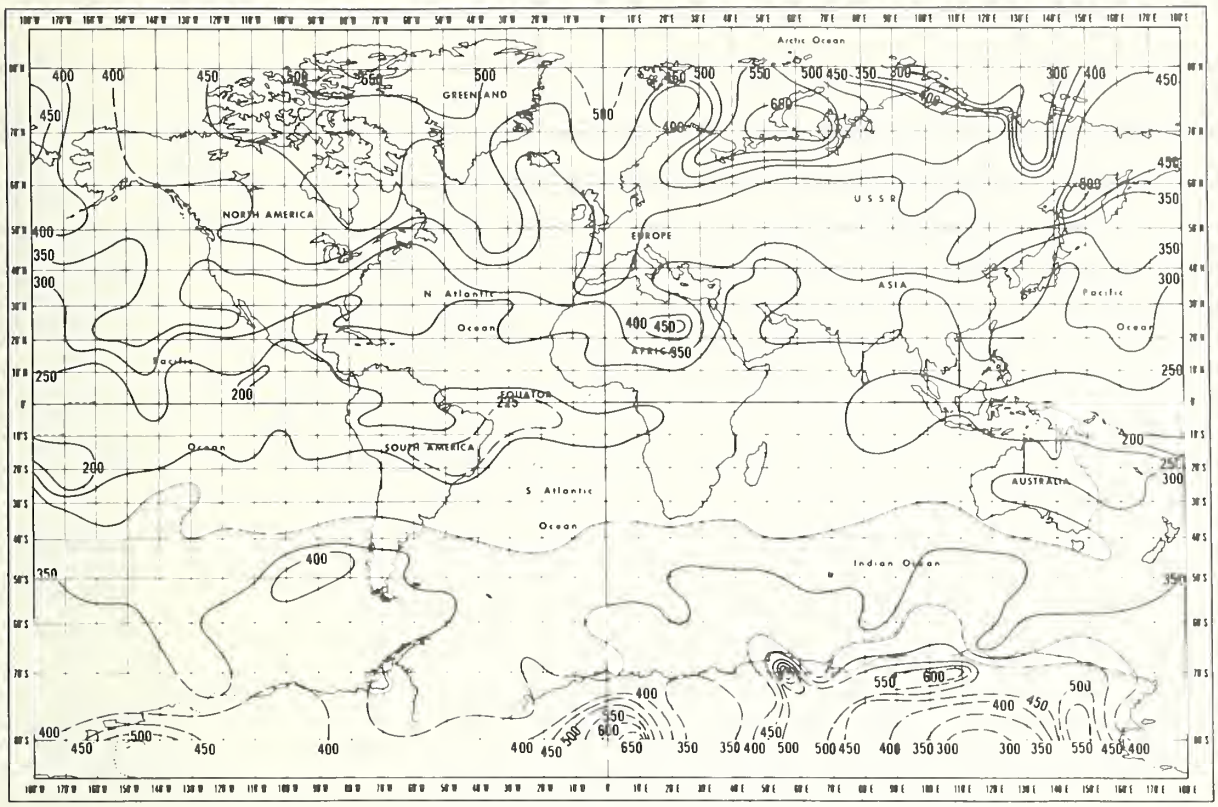


Fig. 5. Atmospheric ozone distribution, worldwide.

atmosphere is well known. Examples of values of solar flux outside the atmosphere are given in Figure 7.

With respect to the motions of the natural stratosphere, questions which may be addressed by CIAP include:

Q.13. "What are the sources of motion within the stratosphere?"

Q.14. "What is the spectrum of motion energy within the stratosphere expressed in terms of mean and standard deviations?"

In Figure 8 is shown a spectrum of wind speed as measured at an altitude of about 100 meters at Caribou, Maine. In this spectrum, showing the square of wind speed as a function of period from 1 minute to 1 year, the peaks appear which are characteristic of the Brunt-Vaisala period of about two minutes, of the 12- and 24-hour tidal motions, of planetary waves of period between 3 and 6 days, and of rotation of the sun (30 days) and of the earth about the sun (1 year). The long-period waves are propagated up from the troposphere more strongly than the short-period waves.

The magnitude of the stratospheric motions has been estimated by Newell in 1969. Table 3 shows that at 20 kilometers the large-scale geostrophic motions, of horizontal scale about 5000 kilometers, have horizontal value of about 10 meters per second in periods of about 30 days. Thermal tidal motions at 20 kilometers having a horizontal scale of about 10,000 kilometers, have horizontal motion of about 0.3 meter per second, and period about 12 hours. The internal gravity waves at 20 kilometers, having a horizontal scale of about 100 kilometers, have a horizontal velocity of about 5 meters per second and a period of about half an hour.

An important question involving theoretical dynamics is:

Q.15. "What is the inter-relation of gradients of temperature, velocity and chemical densities?"

Concerning the dynamic climatology of the natural stratosphere a question is:

Q.16. "What are the mean temperatures and winds in the stratosphere and the variation of these as functions of latitude, longitude and altitude?"

R. E. Newell (1969) has shown the meridional cross section of temperature as a function of altitude and latitude to be as given in Figure 9. In the troposphere, temperatures at the pole are low with respect to the higher temperatures at the equator. On the contrary, in the stratosphere above 20 kilometers, the high temperatures are at the poles and the low temperatures are near the equator.

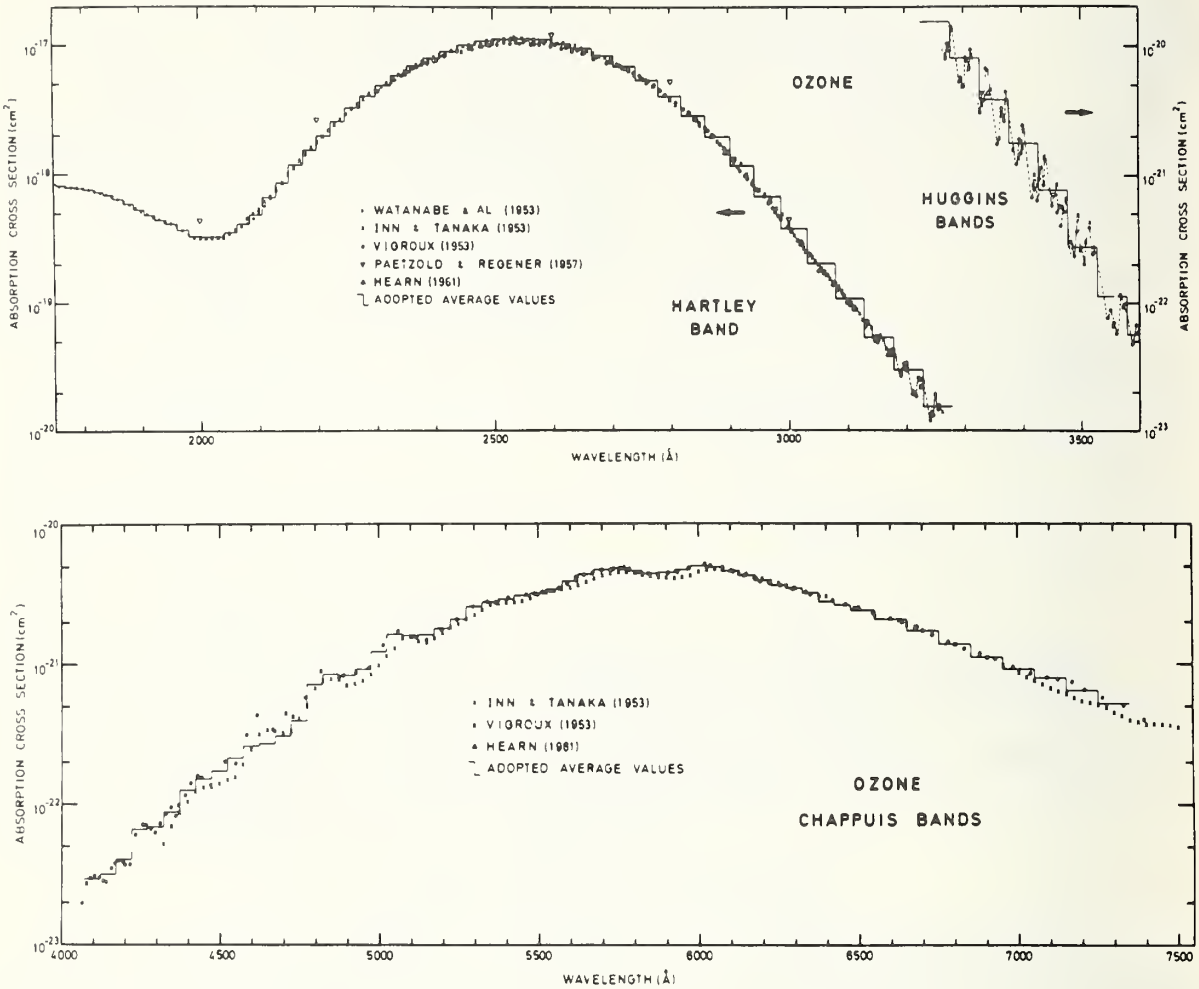


Fig. 6. Ozone absorption cross sections of interest to CIAP (after M. Ackerman, 1971).

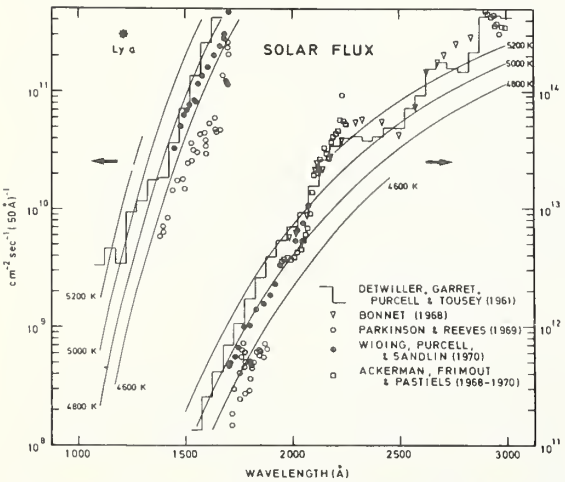


Fig. 7. Solar flux outside the atmosphere (after M. Ackerman, 1971).

In Figure 10, Newell shows the meridional cross section of mean zonal winds. The troposphere cross section illustrates the Hadley cell circulation with hemispheric motions directed by three meridional circulations. By contrast, in the stratosphere, there is a single cell in each of the two hemispheres with effects of meridional circulation moving the heat, as in refrigerator action, from a region of low temperature near the equator to one of higher temperature at the pole.

Shown in Figure 11 are the standard deviations of the meridional wind components and in Figure 12 the relative size of the standard deviations of the ratio of the meridional wind and of the vertical wind. The vertical winds, greatest at about latitude 30 in the troposphere, are approximately 10 times as intense in the stratosphere.

A further question which must be addressed is:

Q.17. "What considerations must be made in simulation of the stratosphere by computer?"

Table 3. Characteristics of atmospheric motions (after R. E. Newell, 1969).

X, Z, horizontal and vertical dimensions *u, w*, horizontal and vertical velocities *t*, time scale

Layer	Large-scale geostrophic motions					Thermal tidal motions				
	X km	*Z* km	*u* m/sec	*w* cm/sec	*t* sec	*X* km	*Z* km	*u* m/sec	*w* cm/sec	*t* sec
Lower thermosphere 100–120 km	10^4	50	30 (1)	5 (2)	5×10^5	10^4	5–50 (3)	30 (1)	5–50 (2)	10^5
Mesopause, 80 km										
Stratopause, 50 km	5×10^3	50	60	1–2 (7, 8)	5×10^5	10^4	20 (9)	10 (9)		10^5
30 km	5×10^3	30	20		5×10^5	10^4	50	0.5 (11)		10^5
20 km	5×10^3	15	10	1	5×10^5	10^4	50	0.3 (11)		10^5
10 km	10^3	15	20	5	5×10^5	10^4	50	0.3 (11)		10^5

Layer	Internal gravity waves					Other				
	X km	*Z* km	*u* m/sec	*w* cm/sec	*t* sec	*X* km	*Z* km	*u* m/sec	*w* cm/sec	*t* sec
Lower thermosphere 100–120 km	10^2	10	50 (4)	50 (2)	10^4 (5)					
Mesopause, 80 km	10–100 (6)		10–100 (6)							
Stratopause, 50 km		2 (12)	6 (10)		10^4 (10)					
30 km		1 (10)	5 (10)		10^4 (10)					
20 km		2 (12)	5 (12)		$2\ 10^3$ (12)					
10 km	20–150 (13)		0.1 (13)		$3\text{–}50 \times 10^2$(13)	Large cumulus clouds 10	10	20	$1\text{–}50 \times 10^2$	10^4

References:
1. Greenhow and Neufeld, 1961.
2. Estimates from $w = uZ/X$ (continuity).
3. Green, 1965.
4. Kochanski, 1966.
5. Hines, 1960.
6. Witt, 1962.
7. Kays and Craig, 1965.
8. Vincent, 1966, private communication.
9. Reed *et al.*, 1966.
10. Newell, Mahoney and Lenhard, 1966.
11. Harris *et al.*, 1962.
12. Weinstein *et al.*, 1966.
13. Gossard, 1962.

Table 4 is significant for its blank spaces summarizing what is presently understood and what is required for CIAP purposes to be known about the stratosphere. Shown at the left are 33 important constituents involved in the 37 most important chemical reactions of 175 considered by Frank Hudson. Of these, about one third are affected photochemically. Also there is about an equal division between hydrogenous, nitrogenous and carboniferous constituents. In the second column is shown the logarithm of the density at 30 kilometers. Only about two thirds of the constituents are listed; the natural vertical distribution of density of the others is unknown.

The sources and sinks of the several constituents are possibly many and various. To be considered are the

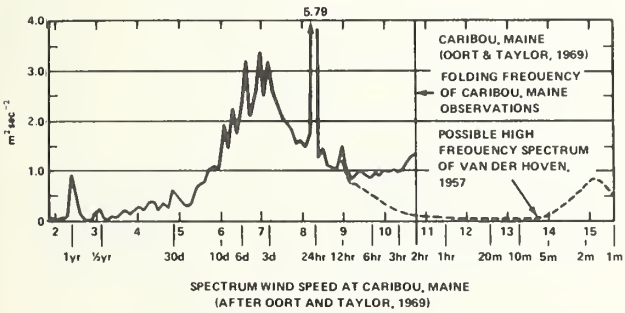


Fig. 8. Spectrum of wind speed at Caribou, Maine.

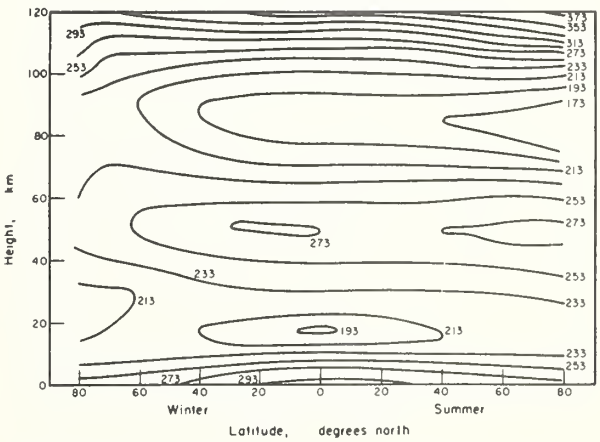


Fig. 9. Meridional cross section of temperature (after R. E. Newell, 1969).

Table 4. Current knowledge of most important stratospheric constituents.

NATURAL STRATOSPHERE						POLLUTION			TRANSPORT				PERTURBATION EFFECT				RADIATION
SPECIES		LOG DENSITY @ 30 km	MEASURE CAPABILITY	SOURCE	SINK	SPECIES	MEASURE CAPABILITY	RATE	ROUTE/ LINE	WAKE	EDDY	CIRCUL	$\bar{n}$	$\sigma(n)$	CLIMATE	PHYSIO-BIO	CRIT WAVE LENGTH nm
O	H	8	*														200 242
O'_0	H	3															242 300
σ	H																450 650
O_2	H	17	*														200 242
O_2O	H	9															242 300
O_2^*	H																450 650
O_3	HJ	12	*									$\overline{n(O_3)v}$					200 300
O_3^*	H																450 650
H_2O	HJC	12	*	THUNDER-STORMS	ABSORPTION	H_2O						$\overline{n(H_2O)v}$					
OH	HJ	7	*			OH											
HO_2	HJ	8	*														
H_2O_2	HJC	10				H_2O_2											
H						H											
N_2	H	17															
N		3															
NO	HJC	10	*	RATE COEFF K_{lm}	RATE COEFF K_{ll}	NO											260 400
NO_2	HJC		*			NO_2											
NO_3	H																
N_2O		12	*			N_2O											
N_2O_5	HJC		*			N_2O_5											500 700
CO_2	H	14	*	TROPOSPHERE	OCEANS	CO_2											
CO	HJ					CO											
CH_4	HJ	12	*		PLANTS	CH_4											
CH_3	H																
CH_2	H		*														
HCHO	H		*			HCHO											
CHO	H																
CH_3O_2	H																
CH_3O	H																
SO_2	H				AOSORPTION	SO_2											
PARTICLES	I			THUNDER STORMS	AOSORPTION												
HNO_3	HC			VOLCANO		HNO_3											<300
HNO_2	J																

REF BORTNER & KUMMLAR 1969 (PLOTTED BY BTL)

TOTAL DENSITY 4 (10) 17

C = CRUTZEN 1971

J = JOHNSTON

H = HUDSON 1971

500 SST IN 1985, (334 x 4) + (166 x 2) ENGINES, 7^{lb}/d INFLIGHT, 120^{lb}/hr NO

1 42 $(10)^6$ ^{lb}/hr NO

J – 58 J – 93

J – 85

2/3 IN NO HEMI, 1/3 SO HEMISPHERE

STARR 1968

CHANGES IN T, $\bar{V}$, PRE C I P, UV FLUX

thunderstorms projecting into the lower stratosphere, the troposphere itself, the oceans, aerosols and volcanic eruptions.

Of the engine emission species, about 15 are currently proposed to be measured by engine testing. Included are the principal constituents: water vapor, nitrogen oxides, carbon oxides and hydrocarbons, sulfur oxides and particles.

The mass of emission species introduced into the stratosphere is dependent upon route projections, frequency of travel on the routes and the rate of emission of engines. One prediction, based on the assumption of 500

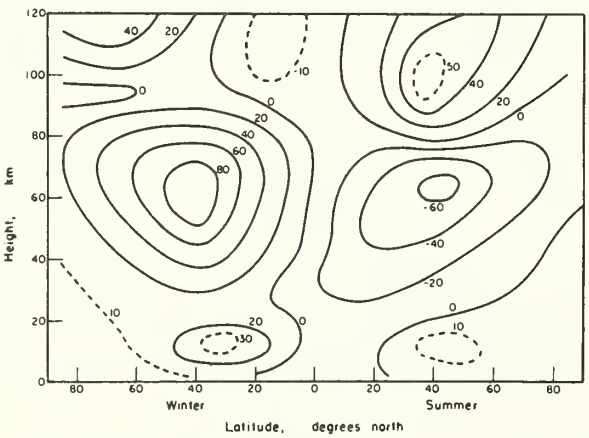


Fig. 10. Meridional cross section of mean zonal wind (after R. E. Newell, 1969).

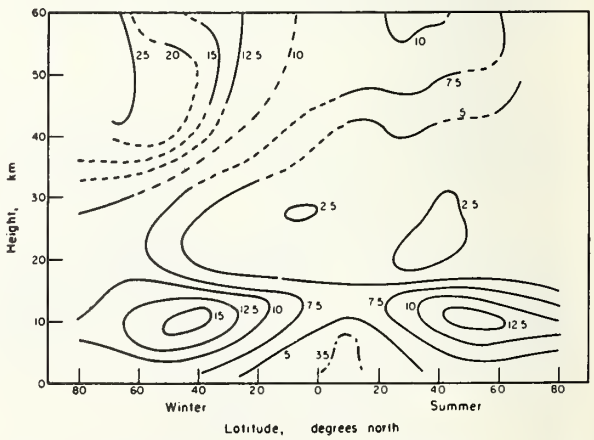


Fig. 11. Meridional cross section of standard deviation of meridional wind component (after R. E. Newell, 1969).

Table 5. Summary of stratospheric computational models.

Model Dimensionality	Examples	Usefulness
Chemical reactions only, no dynamics	U. Washington (Harrison VPI (Levine)	Initial survey of sensitivity of several chemical reactions.
One-dimensional	U. Washington (Harrison) NOAA (Shimazaki) NASA Ames (Whitten) LLL (MacCracken) LMSC (Conti) ARAP (Hilst)	Surveys of sensitivity of chemical reactions using, parameterized diffusive transport. Studies of wake dynamics.
Two-dimensional, with parameterized dynamics	LLL (MacCracken) NOAA (Shimazaki) Institute of Aeronomy (Belgium) (G. Brasseur) LMSC (Conti) ARAP (Hilst)	Effects of diffusive transport on chemistry. Studies of wake dymanics.
Three-dimensional, with dynamics	NCAR-U. Colorado (Washington and London) GFDL (Mahlman) MIT (Cunnold) British Meteorological Office (Murgatroyd)	Studies of dynamics affecting chemistry but not affected by chemistry.
Two-dimensional, with interactive dynamics and chemistry	LLL (MacCracken)	Study of dynamics and chemistry interaction of limited scope.
Three-dimensional, with interaction of dynamics and chemistry	(Post 1974 only)	Complete stratospheric simulation.(Post 1974 only)

Table 6. Basis for computing time estimation.

EQUATION INDICES

DOMAIN	COMPUTATION CYCLE t	t 1	LIMITS
COMPONENTS		j, ℓ	≤42
WAVE BANDS		i,m	≤20
ALTITUDE LEVELS	k	k-1	31

INDICES BY DOMAIN

	NUMBER OF COMPONENTS (j and ℓ)	WAVE BANDS RANGE OF M (m_0 to M)	NUMBER OF WAVE BANDS (i and m)	HEIGHT LAYERS (k)
STRATOSPHERE	18	11 TO 20	10	6
MESOPHERE	30	11 TO 20	10	8
LOWER THERMOSPHERE	42	1 TO 16	16	6
UPPER THERMOSPHERE	30	8 TO 11	4	8
WEIGHTED AVERAGE	30		9	28

TIME, RADIATION COMPUTATION OF, 28 LEVELS	7 75 MS
TIME, RADIATION COMPUTATION OF, 6 LEVELS	1.5 MS
TIME, HYDRODYNAMICS COMPUTATION, TROPOSPHERE	0.18 MS

Table 7. Computer speed comparisons and cost estimates.

CLASS	MANUFACTURER	COMPUTER	RANGE OF RATIO NORMALIZED TO CDC 6600	RATIO AS USED IN REPORT	COST RANGE (IN MILLIONS)	COST USED* IN REPORT (IN MILLIONS)
1	UNIVAC CONTROL DATA UNIVAC	1108 6500 1108 MP	0 3 0 6 0 5 0 8 0 5 0 9	0 4 0 7 0 7	2 4	3
2	CONTROL DATA UNIVAC	6600 1110	1	1	4 6	5
3	CONTROL DATA IBM	7600 360/195	3 8 5 8	5	7 10	8
4	CONTROL DATA TEXAS INST BURROUGHS	STAR ASC-4X ILLIAC IV	22 31 25 35 27 38	30	10 20	12

*BASED ON PRODUCTION QUANTITIES

SST's traveling seven hours per day, emitting at the CONCORDE production rate of about 120 pounds of NO per hour per engine, indicates that the loading into the stratosphere would be of the order of 1.4 million pounds of NO per day. In consideration of the transport processes, one may begin with route projections, which for 1990 may be two thirds in the Northern Hemisphere and one third in the Southern Hemisphere, and which lay down the pollu-

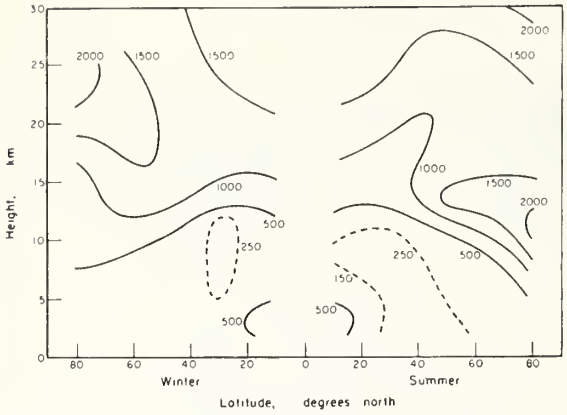


Fig. 12. Meridional cross section of the ratio $\sigma(v)/\sigma(w)$ (after R. E. Newell, 1969).

tants along the vehicle trajectory. This line of effluents is dispersed by the dynamics of the wake, by diffusion due to atmospheric turbulence due to wind shear, and finally by the general circulation of the stratosphere, which moves air masses from one part of the globe to another. A significant effect of turbulence, due to wind shears or temperature gradients, may be the effective alteration of chemical reaction rates from values predicted on the basis of laboratory-determined chemical reaction rate coefficients.

The perturbations of density of species are to be projected by machine computation. In the final column of Table 4 are listed the critical wave bands, expressed in nanometers, of the photochemically active radiation affecting distribution of the constituents.

Figure 13 is a hypothetical enumeration which suggests there are large differences in the relative contribution of several terms in the differential equations whose integration simultaneously for about 33 different species provides the basis for establishing the equilibrium of the species at each altitude. By substituting typical values for reactions involving NO and O_3 at 20 kilometers, the first term of the contribution (that is, the photochemical term) has a mean value of approximately 10^9. This value may be compared with the reactive chemistry term (the product of the rate coefficient and the contributing mean densities) which has a value of about 10. One may see that in the daytime the photochemistry dominates the reactive chemistry.

The chemical "Loss" terms may be considered to be comparable in size with the chemical "Production" terms.

The advection term involving the wind velocity and the gradient of density is seen in this instance to have a value of about 10^6. Although in the daytime this term is swamped by the photochemistry, at nighttime the advection term swamps the chemical reactivity. A third term, for diffusion (important principally for transport in the vertical dimension), has a value of about 10.

The ratio of standard deviation to mean value, of these terms, is also indicated in the second row. The photochemical term uncertainty is about 10 if the solar flux activating the photochemistry is measured in situ, but is larger than about 10^3 or more if the flux is estimated from an out-of-the-atmosphere measurement diminished by an exponential factor dependent upon the integrated densities in the intervening column of absorbing constituents. Using the ozone column variation of Prabhakara, the photochemically active solar flux at 20 kilometers may be uncertain by a factor of 10^3 to 10^5. The uncertainty of the chemical rate coefficient in some instances is 10^3; however

$$\frac{\partial n_i}{\partial t} = \left[\left\{ J_k(z) = \int_{\lambda_1}^{\lambda_2} \sigma(\lambda)\epsilon(\lambda) I(\lambda, z)\, d\lambda = \int_{\lambda_1}^{\lambda_2} \sigma(\lambda)\epsilon(\lambda) I(\lambda, \infty)\, e^{-\sum_i \sigma_i(\lambda) \int_z^\infty n_i \sec x\, dz}\, d\lambda \right\} \bar{n}_k + k_{jk} \left\{ \bar{n}_j \bar{n}_k + \overline{n_j' n_k'} \right\}\right] - [\text{Loss}] - \bar{V} \cdot \nabla n_i - D\nabla^2 n_i$$

Mean Value: $\underbrace{10^{-18} \cdot 1 \cdot 10^{15} \cdot 10^{12}}_{10^9}$ $\quad \underbrace{10^{-23} \cdot 10^{12} \cdot 10^{12}}_{10}$ $\quad \underbrace{10^4 \cdot 10^2}_{10^6}$ $\quad \underbrace{}_{10}$

S.D./Mean $\underbrace{10 + 10 + 10}_{10}$ in situ $\quad \underbrace{10 + 10 + 10 \quad + 10^3}_{10^3}$ computed $\quad \underbrace{10^3 + 10 + 10}_{10^3}$ $\quad \underbrace{10^3 + 10^3}_{10^3}$ $\quad \underbrace{10^6}_{10^6}$

(j) + (k) → (i) + (m)

n_i Density distribution, for each altitude, determined by integrating simultaneously 37 equations of the form above, relating 33 different species

σ Absorption cross section

ϵ Quantum yield

I Solar flux, of wavelength λ and at altitude z

x Solar zenith angle

k Reaction rate coefficient

$\bar{V}$ Mean wind velocity

∇n_i Gradient of the density

D Diffusion coefficient

$\nabla^2 n_i$ Laplacian of density, representing a source or sink

$\overline{n_i' n_k'}$ Cross correlation of density fluctuations

Fig. 13. Number continuity equation.

Table 8. 1973-1975 environmental model computation times.

(1)	(2)	(3)	(4)	(5)	(6)	(7)	(8)	(9)	(10)	(11)	(12)	(13)	(14)
MODEL	MODEL SUBDIVISIONS	NUMBER HORIZONTAL POINTS	NUMBER VERTICAL POINTS	TOTAL POINTS	TIME STEP	PROGNOSIS PERIOD	NUMBER TIME STEPS	TOTAL POINT CALCULATIONS (5) X (8)	ESTIMATED CDC 6600 TIME PER POINT	TOTAL 6600 TIME PER RUN	NUMBER OF RUNS PER DAY	TOTAL 6600 TIME PER DAY	SECONDS PER SECOND
TROPO/ STRATO/ MESO/ THERMO-SPHERE	U, V, T, P, n_i	8000	33	264,000	10 MIN	36 HR	216	57×10^6	1.0 MS	60 HR	4	240 HR	10
	RADIATION, ETC.	8000	28	224,000	2 HR	36 HR	18	4×10^6	40 MS				

in most instances rate coefficients are known within a factor of 10. Contributing to the uncertainty of the advection term are the uncertainties of winds and of the gradients, each about 10^3. The uncertainty of the diffusion term has a contribution from the Laplacian of the density (representing the sources and sinks) and from the diffusion coefficient. The diffusion term uncertainty is likely to be of the order of 10^6.

This over-simplified expression suggests that a sensitivity analysis is needed to determine which sources of uncertainty in computing the density-height distribution of the perturbed stratosphere are more sensitive in contributing to the uncertainty of the final result. Conclusions to be drawn from the example indicate: (1) that determination of densities of as many of these constituents as possible should be made simultaneously in space and time; (2) that the photochemically reactive solar ultraviolet flux should also be determined at the same time and location; (3) that determination of velocities and diffusion transport also require emphasis; and (4) that there is a premium for determinations, such as satellite measurements, that yield a field of densities, from which gradients and Laplacian values may be determined.

Other questions which relate to simulation of the stratosphere by computer include:

Q.18. "What are expected limitations of stratospheric simulation to be done in the next two years?"

Q.19. "What abridgements of a complete model of the stratosphere-troposphere-ocean, as planned for future development at Global Fluid Dynamics Laboratory, may seem appropriate to derive answers desired by CIAP?"

In a review of computational models useful for analysis and simulation of the stratosphere, the DOT Workshop on Computational Modeling of the Stratosphere (April 1972, Asilomar, California) summarized the types of models in a manner given in Table 5. At the top of the list of Table 5 are the simplest modeling efforts primarily useful for making estimates of chemical reactions without consideration of the dynamics. At the bottom of the list, ordered in degree of complication, are the elaborate and computer-time-consuming models operating with three space dimensions and time dependence for studies of dynamics which affect the chemistry but are not in turn affected by the chemistry. The most elaborate modeling effort (i.e. the three-dimensional models which take into account the mutual interaction of dynamics and chemistry) are envisioned at the Global Fluid Dynamics Laboratory (GFDL) and at UCLA but are not expected to simulate the stratosphere successfully until some time after 1974. The tabulation in Table 5 shows that a hierarchy of models may be used to address different aspects of the problem; none of them may be expected to accomplish complete simulation by 1974; a 1974 CIAP report must depend, therefore, on estimates based on incomplete simulation.

The complete simulation of the stratosphere, in reaction with the troposphere below and the mesosphere above, is a difficult problem but quite within the range of computer capacity existing today. In Figure 14 are shown some of the digital numerical relations used in estimates of computing time required for a simulation of the upper atmosphere up to an altitude of about 300 kilometers. The term S_l is representative of the photochemical J term given in Figure 13. The subscript l denotes a constituent, m denotes a wave band and k denotes height level. The relation for n_l describes the solution of l simultaneous equations constituted of the number density equations; $N_j(k)$ denotes the height-integrated column of densities. Q represents the heating. The equations of Figure 14 were used to estimate the time for computation.

The equation indices, given in Table 6, are enumerated for each of the major domains of the upper atmosphere. On the basis of these numbers and by comparison with the computer time spent in executing the hydrodynamic computations of Leith's model of the troposphere, the time for computing the radiation effects in the upper atmosphere is estimated and given also in Table 6. For each grid point of Leith's model of the troposphere, 0.18 milliseconds is required for computation of tropospheric dynamics. The computation for the radiation effects, using the numerical relations of Figure 14 for six levels is 1.5 milliseconds and for 28 levels 7.5 milliseconds. The time for computation of the radiation at each grid point is seen to be roughly 40 times the time spent in computing the hydrodynamics for that grid point.

The computations described above are estimates based on performance of the Lawrence Livermore Laboratory computer CDC 6600. There will exist by 1973 at least three computers of computational capacity at least 30 times that of the CDC 6600 with which they are compared in Table 7.

$$S_\ell = \frac{1}{\mathbb{P}_\ell} \sum_{m=m_0}^{M} \underbrace{\sigma_m \ell \left\{ I_m \exp\left[F(X) \sum_i \sigma_{\ell,i} N_i(k-1) \right] \right\}}_{J_{\ell,m}}$$

Wherein. ℓ CONSTITUENT; m WAVE BAND; k HEIGHT LEVEL

$$n_\ell = \left[\begin{array}{l} \text{SOLUTION OF } \ell \times \ell \\ \text{SIMULTANEOUS} \\ \text{EQUATIONS} \end{array} \right]$$

$$N_i(k) = N_i(k-1) \quad n_i(k-1)$$

WHERE $n_\ell(k)$ ARE THE RESULTS OF THE SOLUTION OF THE $\ell \times \ell$ SIMULTANEOUS EQUATION SET (SEE EQUATION 37 OF THIS APPENDIX)

$$O^{i+1} = O^i + \Delta t \left[\sum_\ell n_\ell \sum_m J_{\ell,m} \right]$$

Fig. 14. Digital numerical relations used in atmospheric simulation.

These machines are the CDC STAR at Lawrence Livermore Laboratory, the Texas Instrument Co. ASC-4X computer at the Global Fluid Dynamics Laboratory, and the Burroughs Illiac IV at the NASA Ames Laboratory. In terms of the problem mentioned in these remarks and the capacity of the machines available in the 1973 to 1975 period, the estimates of overall computer time are as given in Table 8. The computation of both hydrodynamics and radiation for the atmosphere from sea level to 300 kilometers using 28 levels above the troposphere and 8000 horizontal grid points may be expected to require 60 hours of CDC 6600 computer time or two hours of a computer like the CDC STAR, Illiac IV or the Texas Inst. Co. ASC-4X.

With respect to instrumenting the measurement of constituents and of transport within the natural stratosphere, a question for CIAP is:

Q.20. "What concepts and techniques for remote and in situ measurements are presently available and most applicable to measure water vapor, ozone, the trace gases important to CIAP, and particulates?"

Some of the experimental methods in exploring the atmosphere have been described by Difedele, 1968, as given in Figure 15. One sees that in the region 20 to 40 kilometers the principal means are aircraft and balloons doing meteorologic sounding, ground-based Lidar, and various means of determining absorption of sunlight and the infrared thermal emission.

In examining the changes in the troposphere which may be consequent on the perturbation of the stratosphere, there are questions to be addressed by CIAP which are:

Q.21. "What is known of the climatology of the troposphere by direct observation as it would relate to the troposphere-stratosphere relations?"

Q.22. "How may the effects of changes in radiation heating of the troposphere resulting from changes in constituent density distribution within the stratosphere be interpreted in terms of altitudes and directions of mean winds (at 500 millibars, say) and of precipitation in the short term and in the long term?"

Some suggestions as to the form of the answers are given by a study ten years ago by Professor Yale Mintz involving a simple two-layer tropospheric model. The model does not take into account the important influence of geography.

First of all, the cellular meridional circulation on a rotating earth, according to a picture attributed to G. C.

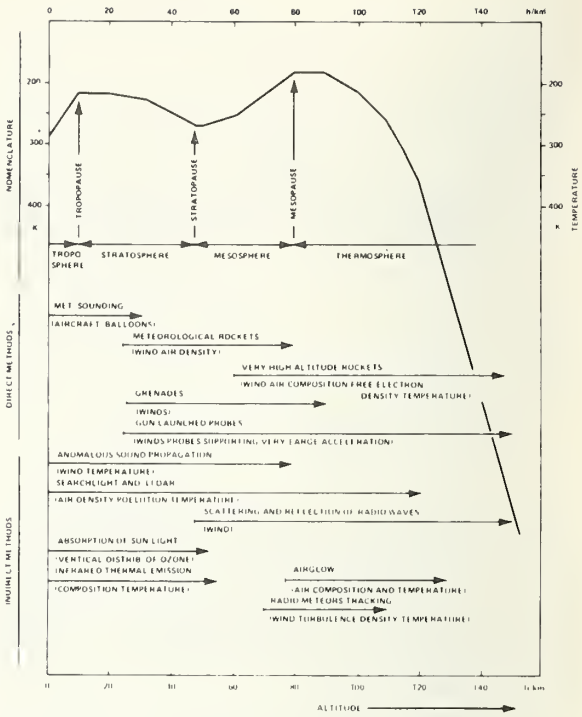


Fig. 15. Experimental methods in exploring the atmosphere.

Rossby in 1945, is shown in Figure 16. In this figure are represented the three Hadley-Ferrell cells which produce low-level easterly winds in the lower tropics, westerly winds in the mid-latitudes, and easterly winds in the polar regions. The boundaries between the oppositely directed winds are the source of inertial waves which, as in Figure 16, give a roughly sinusoidal form to the zonal wind trajectories as they circle the earth. Yale Mintz in his study found that this wave form was necessary for the transport of heat in the troposphere from the high temperatures of the equator to the low temperatures of the poles.

Figure 17 shows a polar view of the zonal wave motion enhancing the meridional heat transport, and also the relations of the underlying high-pressure and low-pressure areas. The circulation in the northern hemisphere is counter-clockwise around the low-pressure areas and clockwise around the high-pressure areas. The number of the nodes of this wave motion of circulation is a function of the heating and temperature gradient.

In Figure 18 is shown the model Mintz used to describe the radiation budget for the planet earth which depends on infrared emission from carbon dioxide within the troposphere, and on the solarization as it is affected by the troposphere and surface albedo. There is a net differential heating (ΔQ) which is the difference between these values. Employing this model Yale Mintz derived a change in heating (expressed in kilojoules per second) between summer and winter of 2×10^{12} to 8×10^{12} as shown in Figure 19. The planetary wave number, that is, the representative number of nodes of the mid-latitude zonal trajectory around the earth, is described as a function inversely proportional to the heating of the upper troposphere. He found the nodes to number from 7 to 8 in the

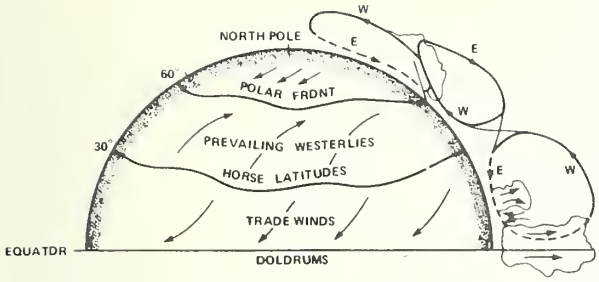
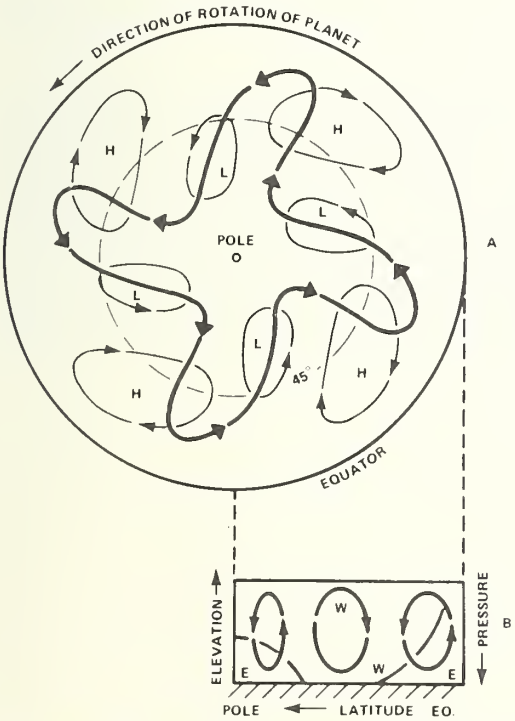


Fig. 16. Cellular meridional circulation of a rotating earth.

summer and 5 to 6 in the winter. Mintz suggested the root-mean-square velocity of the mean hemispheric wind is a function of the heating differential divided by the temperature in the upper troposphere, and by this method of estimation calculated that the wind speeds in the winter are approximately twice that of the summer as has been observed.

A sketch in Figure 19 suggests some implications. W indicates a schematic trajectory of winds in the troposphere near the polar front in the wintertime under the assumption that the shape is dominantly positioned by the persistent high-pressure region of continental Siberia. S indicates the summer winds trajectory with a slight shift of phase within the continental United States in the summertime. Both of the summer and winter winds, from the standpoint of the United States seem to originate in Alberta, bringing dry polar air down through the west until reaching the southernmost extreme at about Oklahoma. The zonal trajectory there turns north, carrying with it masses of moist tropical air dragged out of the Gulf stream alternating with masses of dry polar air. A consequence of this process is that the United States west of the Mississippi is generally dry in contrast with the east of the Mississippi which is more humid.

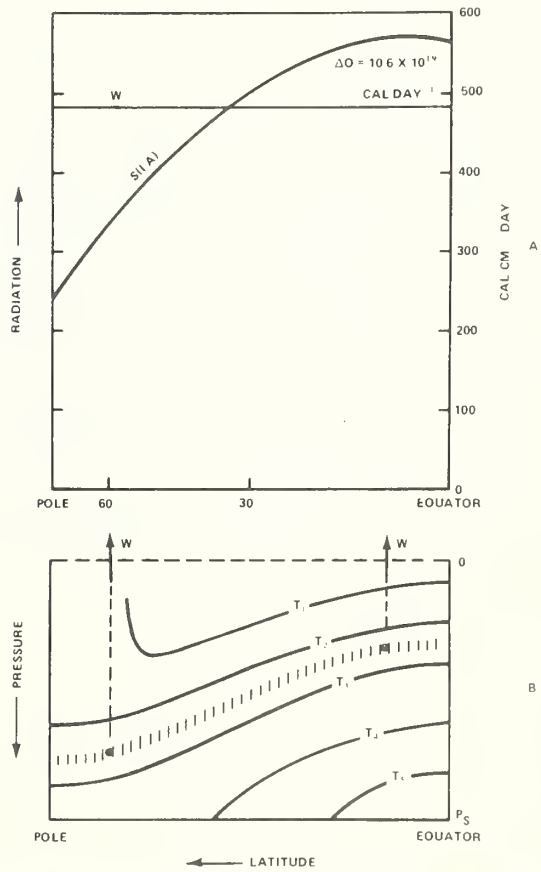
A possible effect of a markedly cooler solarization of the stratosphere resulting from changes in the stratosphere may be speculated. Pursuing the suggestion drawn from Mintz' study, the winds may be stronger, and there may be a phase shift of the zonal wave. This may bring, as this sketch suggests, warm humid moist air onto the shores of the Pacific Northwest with the polar dry air coming down into the Eastern Part of the United States. Such an effect, if



A) STREAMLINES OF THE FLOW AT THE MIDOLE AND UPPER LEVELS (HEAVY LINE) AND NEAR THE GROUND (THIN LINE). L = LOW PRESSURE, H = HIGH PRESSURE.

B) CROSS SECTION SHOWING THE ZONALLY-AVERAGED MERIDIONAL CIRCULATION, AND THE ZONALLY-AVERAGED ZONAL WIND, WHERE W = WESTERLY WIND, E = EASTERLY WIND

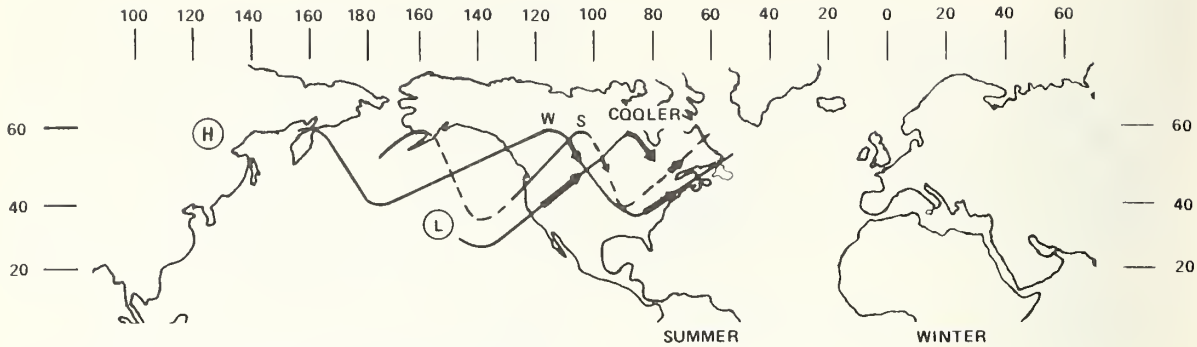
Fig. 17. A wave regime of general circulation (after Mintz, 1961).



A ENERGY GAIN AND LOSS AS A FUNCTION OF LATITUOE, WHERE S IS THE INSOLATION PER UNIT HORIZDNTAL AREA PER UNIT TIME, A IS THE PLANETARY ALBEDO, W IS THE INFRARED EMISSION TO SPACE, AND ΔO IS THE NET OIFFERENTIAL HEATING

B. INFRAREO EMISSION TD SPACE WHEN THE RELATIVE HUMIOITY IS CONSTANT ALONG THE ISDTHERMAL SURFACES T THE SHAOED BAND REPRESENTS A LAYER OF WATER VAPOR, OF CONSTANT OPTICAL THICKNESS, WHICH CONTRIBUTES TD THE TOTAL INFRAREO EMISSION W

Fig. 18. Model of the radiation budget for the planet earth.



	SUMMER	WINTER
CHANGE IN HEATING ΔQ (KJ sec^{-1})	$2(10)^{12}$	$8(10)^{12}$
PLANETARY WAVE NUMBER $n_d = \frac{a \Omega m}{R} 2^{1/4} \left(\frac{C_p}{T_z s}\right)^{1/2}$	7.8	5.6
MEAN SQUARE VELOCITY $(\overline{V_z^2})^{1/2} \cong \left[\frac{q\, m\, 2^{1/4}\, \Delta Q}{\pi\, a\, p_s \sin 30\, (C_p T_z S)^{1/2}}\right]^{1/2}$	X 1	X 2

Fig. 19. Implications of climatic change.

realized, would strongly affect the U.S. climate. These speculations may be tested by climatological simulation.

A panel of the National Academy of Sciences (Prof. K. Smith Jr., Chairman) reviewed in October 1971 the effect of the ultraviolet radiation in the troposphere on man, animals and plants. Increased penetration of the ultraviolet into the atmosphere may be a possible effect of diminution of the ozone column within the stratosphere. Certainly questions for CIAP are:

Q.23. "What are the effects of ultraviolet radiation on living tissue, in terms of mean and standard deviations?"

Q.24. "What wave bands of radiation are particularly effective for damage?"

Q.25. "What is now known as to the levels of radiation intensity felt by living tissue in the dangerous wave bands?"

Some answers to these questions are given in H. Blum "Carcinogenesis by Ultraviolet Light," Princeton University Press, 1959.

As a final conclusion of the study it is desirable that the physiological-biological-botanical effects, as the true impacts that people may realize, be expressed to the extent possible in terms of economic measures. Expression by economic measure is advised both for the reason that an economic measure is an additional index by which the significance of a technical statistic may be appreciated, and for the reason that it provides a common unit of comparison with results of other studies which are not to be done as part of the CIAP program but which may represent the benefits, for example, of vehicles traveling in the stratosphere.

-- NOTES --

-- NOTES --

APPENDIX C

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CIAP COMPUTATIONAL MODELING WORKSHOP
ASILOMAR, PACIFIC GROVE, CALIFORNIA

11-14 April 1972

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